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Small Scale Decontamination of Radioactive Water by Ion Exchange
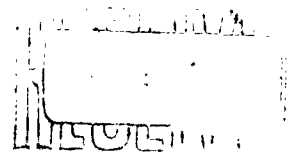
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A Report on

SMALL-SCALE DECONTAMINATION OF RADIOACTIVE WATERS BY ION EXCHANGE

Prepared for

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ABSTRACT

Immediately following nuclear war events, it is anticipated that surface waters will be contaminated by radioactive fallout materials and it appears desirable to minimize the amounts of ingested radioactive materials for a period of time to reduce the radiation doses of the population. Although drinking waters free of radioactivity may be provided by prior storage, or decontamination may be achieved to an acceptable degree by emergency means utilizing materials common to most households, there is still an apparent need for a small-scale commercial decontamination unit.

In order to supply an appropriate basis for design of such small-scale commercial units, it has been necessary to determine the chemical and physical natures of the radiocontaminants, to review the various methods available for decontamination and their respective performance in laboratory and field experiments, and to select suitable decontamination methods in relation to the anticipated problem. The basic design criteria are those of simplicity, of capability, and of reasonably low cost; the ion exchange process is apparently most appropriate for this purpose.

Laboratory studies have shown that turbidity or suspended solids, particularly clay minerals in the raw waters impose a limitation on the ion exchange process, and it is desirable to require prior filtration of the raw waters. Specific laboratory studies were conducted to determine the relationship between suspended solids and ion exchange resins and to ascertain the effect of suspended solids on the removal of radiostrontium by ion exchange resins.

A decontamination unit which includes both filtration and ion exchange, using synthetic ion exchange resins, is specified, and considerations in the design of the unit are enumerated. A decontamination unit meeting the prescribed criteria would be reasonably simple in operation and would be capable of adequate decontamination performance, however it appears that the cost of the unit would be greater than desired. A detailed study of the relative costs of the decontamination unit and alternate sources of uncontaminated waters, such as emergency stores of water, is desirable

under the circumstances. Preliminary estimates indicate the decontamination unit would not be more economical than stored water unless the total volume of required water exceeded two or three hundred gallons.

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INTRODUCTION

That portion of the population surviving the immediate effects of blast,
fire and initial radiation accompanying nuclear ware events would be faced with
the hazards associated with residual or fallout radiation. The acute physiological
effects from fallout radiation experienced by the total population are difficult
to predict except in a general sense because of variations in fallout deposition,
in shelter adequacy, and in individual radiation damage resistance. It appears
probable that an acute effects spectrum would be encountered, ranging from
negligible damage for those adequately sheltered in areas of low fallout to
the fatalities associated with insufficient shelter provisions in high radiation
fields. The consumption of waters containing radioactivity during the postattack
phase could contribute toward the radiation doses received and the resulting acute
effects; presumably it would be beneficial if the waters consumed during this
period did not contain radioactive material.

It is desirable to review some concepts and available methods relating to
the removal of radioactive contaminants from water supplies. The problems as
they concern small-scale emergency water supply are emphasized; water supplies
for larger populations, perhaps greater than ten people, are considered to be
outside the scope of this report.

Decontamination Considerations

To what degree should drinking waters be decontaminated during the postattack
phase of a nuclear war? Although there is no simple answer to this question,
estimates of the required decontamination might be calculated from the probable
contamination level and the concentration considered to be acceptable. Even a
cursory review of the variability in probable fallout deposition and subsequent
surface water contamination indicates that the specification of a given degree of
required decontamination as being universally applicable is unlikely.

It is believed that the human body can accommodate certain radiation doses and dose rates such that, on the average, adverse physiological effects are not demonstrable; this concept is the basis for the current directives promulgated to regulate peace time radiation exposures of the public. The different relative hazards posed by the individual radionuclide species in the environment are recognized in the current peace time regulations adopted by the Atomic Energy Commission (1). An uncontrolled large-scale release of mixed fission products as a result of nuclear war events would present a more complicated problem and directives concerning maximum permissible emergency concentrations of radioactivity in drinking waters necessarily must be broad in nature until accumulated data make more certain the probable relative concentrations of specific radionuclides and their related biological effects. Decisions regarding acceptable levels of biological damage to the general population are considered prerequisite to the establishment of maximum permissible concentrations.

Two approaches toward delineation of maximum permissible emergency concentrations of radionuclides in drinking water have been proposed. Because the more hazardous radionuclides become of greater relative significance as decay reduces the total radioactivity of fallout materials, Morgan and Straub (3) suggested a limit decreasing with time,

$$MPC = 10^{-3} \ t^{-1.2} \tag{1}$$

where MPC is the maximum permissible emergency concentration, $\mu c/m.$, and t is the time in days following the explosion.

Studies such as those by Kaufman and Dennins (4) and by Hawkins (5) indicate and discuss the many complex factors involved in water contamination by radioactive

materials. Although these and similar studies demonstrate that accurate estimations
of contamination are not to be obtained by simple means, detailed studies of
specific watersheds under various assumed attack conditions conceiveably could
result in approximate contamination values. For example, preliminary data made
available by Grune (6), studying the water contamination of a certain New England
watershed as a result of a single 5 MT weapon explosion some 50 miles away, suggest
an average decontamination factor of from 3 to 30 might be required for the first
ten days. This is of course but a single example; studies of other watersheds
with related anticipated attack conditions possibly would result in either higher
or lower estimates of the required degree of decontamination.

Recognition of the external radiation field and the available shelter
protection should be made when considering the degree of water contamination
and the attendent required decontamination. Hawkins (7) compared the internal
and the external doses when a sheltered population consumes water from a
distribution reservoir contaminated with fallout materials. Under the assumed
conditions the thyroid was determined to be the controlling organ and radioiodine,
from 10% soluble fallout particles and assumed to be 100% soluble in the stomach,
was taken as the sole contributor to the thyroid dose. Some of Hawkins' calculated
results are shown in Figure 1; the relative importance of contaminated water
consumption apparently is greater under circumstances where the shelter is of
high effectiveness, i.e. at high values of shelter protection factor. On the
other hand, when the available shelter protection is relatively small, consumption
of contaminated water appears to be of less relative importance.

Another factor which might be considered is the radiation field around a
water decontamination unit. If contaminated water is brought into a shelter area

where it is processed to remove the radiocontaminants before consumption, the accumulation of radioactive materials in the decontamination unit could result in a source of additional whole body dose although it generally can be assumed to be insignificant.

Delay in the consumption of contaminated waters appears to be primarily beneficial in relation to acute effects, and the delay may be achieved by use of uncontaminated waters or by employment of a decontamination device. If long-term effects are considered, delay in contaminated water consumption appears to be of little benefit except where the largest concentrations of those radionuclides associated with life span shortening occur during the delay period.

5

Committed Thyroid Dose, rem.
External-Dose, rem.

$\cdot 10^1$

1.0

10^{-1}

10^{-2}

10^{-3}

10^{-4}

Consumption of contaminated
water starting at

2 days
5 days
10 days
20 days

After Hawkins (5)

10^1 10^2 10^3 10^4

Shelter Protection Factor
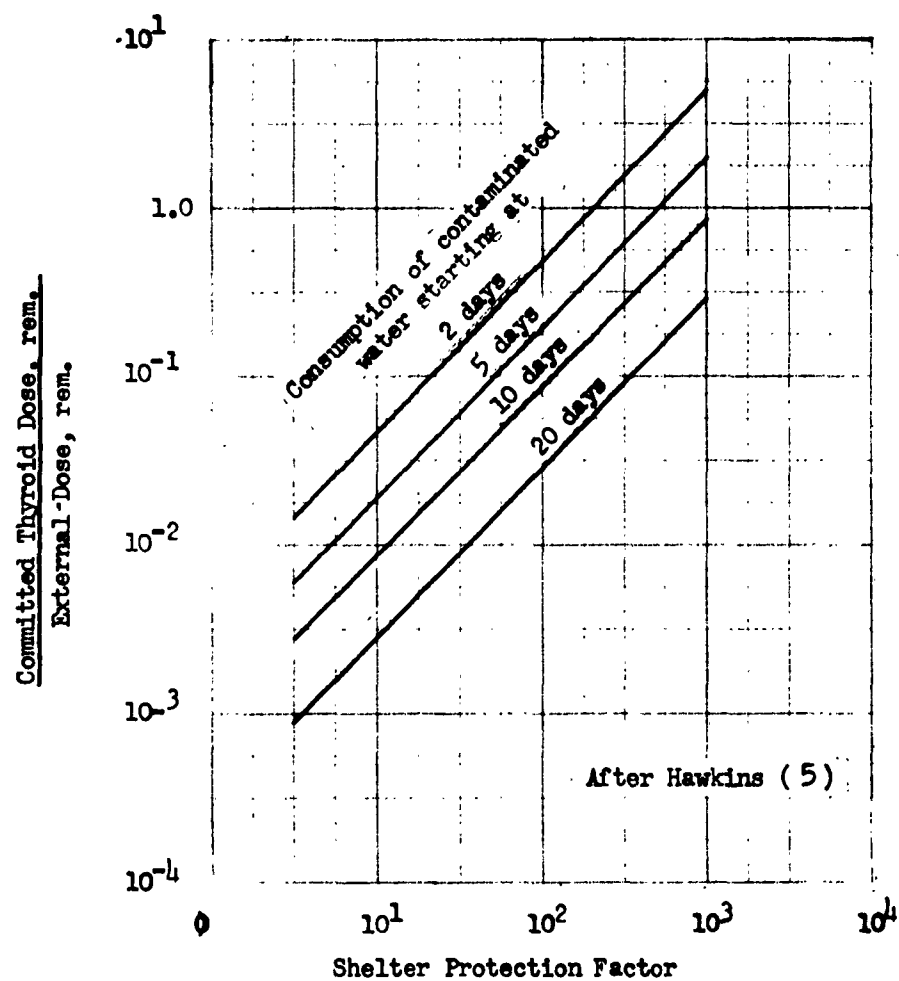
FIGURE 1.   SIGNIFICANCE OF INTERNAL DOSE IN

RELATION TO SELECTED VARIABLES.

Decontamination Methods

Considerable effort has been made toward the evaluation of the various possible decontamination methods for radioactive waters, and much of this work has been accomplished and reported as a joint program between the Health Physics Division, Oak Ridge National Laboratory, and the Robert A. Taft Sanitary Engineering Center, Public Health Center (8). Interest has centered upon the methods available in conventional water treatment plants, and these include conventional coagulation, sand filtration, and water softening, including ion exchange. Other methods investigated include phosphate coagulation, electrodialysis with permselective membranes, distillation, and procedures based upon the addition of either metallic dusts or clay minerals.

The significance of previous decontamination studies can best be determined in relation to the nature of the anticipated water radiocontaminants following a nuclear attack. Lacy (9) has discussed the general characteristics of fallout materials, and studies such as those reported by Lowe et al. (10) and by Lindsten et al. (11) indicate that radionuclides associated with the fallout particulates are but sparingly soluble in water. A general value for solubility of ten percent has been suggested as a conservative upper limit, and the radionuclides of strontium, iodine, and cesium are found in the water soluble group.

Since the larger portion of the radioactive materials associated with fallout is insoluble, laboratory results obtained with reactor-produced radionuclides and simulated waters are not directly applicable to waters containing radioactive fallout materials. However, these results are useful in predicting, at least approximately, the behavior of the soluble fractions of the fallout materials.

Evaluations of the effectiveness of the conventional water treatment processes have been made in some cases by observing the passage of nuclear test fallout

through actual plants. These studies, such as that by Bell et al. (12) indicate removals ranging from 35% to 80% for coagulation, sedimentation, and filtration. In some cases the low concentrations of fallout made measurements difficult, but it appears that conventional water treatment processes cannot provide decontamination factors greater than 5 to 10.

In relation to the scope of this report, probably the most significant information regarding decontamination methods has been provided by the U. S. Army Engineer Research and Development Laboratory (10) (13). In these studies actual bomb debris, i.e. pan collected material in the vicinity of the explosion, was used to approximate fallout materials, and it is believed that the experimental results may be extrapolated to nuclear war conditions. These studies concluded that 'complete' decontamination was possible through a combination of a primary clarification step, such as coagulation and sedimentation or as filtration, with a secondary treatment of distillation or of ion exchange. Although the clarification step provided a greater removal of gross radioactivity, it is possible that the secondary step is of greater importance in relation to committed internal dose.

The primary criteria which a small-scale water decontamination unit must satisfy are adequate performance, simplicity of operation and reasonably low cost. It is difficult, and perhaps meaningless, to assign priority to these criteria; a proposed unit should satisfy all criteria to be fully acceptable. To be successful, coagulation requires special training and experience on the part of the operator and distillation is not possible without a source of heat. From considerations such as these it appears that a decontamination unit incorporating filtration and ion exchange is perhaps best suited for the removal of radioactive fallout materials from small volumes of drinking water.

Such a decontamination unit would presumably be capable of achieving decontamination factors considerably greater than those considered necessary to

reduce the probable contamination levels to the maximum permissible emergency concentrations, but this is in the proper direction to err. There are two reasons for 'overdesign' of the units; despite the best efforts to predict probable levels of contamination, there will probably be some locations which will experience a significantly greater problem than anticipated, and there is the possibility that maximum permissible emergency concentrations would be revised downward in the future if the currently acceptable level of biological damage to the public is felt to be excessive in relation to decontamination effectiveness available.

If a combination of filtration and ion exchange processes is considered satisfactory for decontamination, attempts to improve either the simplicity of operation or the cost of the unit should be directed toward investigating the processes individually. Since filtration is ineffective in removing the soluble fractions of radioactive materials, and since the radionuclides of strontium, cesium, and iodine, generally considered to be the most hazardous from an ingestion viewpoint, are among the water soluble group, filtration does not appear to be satisfactory without secondary treatment.

Insufficient information is available to determine the feasibility of using ion exchange without prior filtration of the influent waters. Passage of unfiltered water through an ion exchange bed conceiveably could (a) remove the suspended solids, thereby resulting in a decreased exchange capacity by blocking resin pores or exchange sites or in hindering the rate of flow through the bed, or (b) permit the suspended solids to pass through the bed which, since radioactive materials could be incorporated with the suspended solids, would lead to a decreased decontamination factor. The need to investigate the interaction between suspended solids and ion exchange resins became the motivation for the present study.

Scope of Study

The primary objective of this study is the delineation of a suitable basis for design of ion exchange systems for small-scale decontamination of drinking water following a nuclear attack. Emphasis is placed on those radionuclides present in fallout materials which are of health significance.

The chemical and physical states of the radionuclides are considered in relation to the theoretical concepts of the ion exchange process and the properties of the exchange media. Elementary ion exchange theory is presented to the extent necessary in meeting the primary objective, and information regarding the chemical and physical states of the radionuclides is based upon available published material.

Since the removal of radionuclides from contaminated waters by ion exchange is strongly influenced, if not dictated, by the characteristics of the waters themselves, a brief survey of the variations in total hardness and turbidity to be found in the surface waters of the United States is presented. The role of suspended solids in limiting the degree of decontamination achieved by the ion exchange process is indicated and briefly discussed.

A review of filtration theory, as it applies to beds of granular solids, is presented as a basis of understanding possible interactions between suspended solids and the resin particles of an ion exchange bed.

Laboratory investigations of suspended solids-resin interactions are reported, and experimental results are tentatively expressed by theoretical equations. Most of the investigation dealt with suspended solids from local soil samples which contained a high percentage of quartz and the predominate clay mineral was that of kaolinite; studies were also made with standard or reference clay minerals to verify the results obtained with local soils.

Additional laboratory studies indicating the behavior of radionuclides, particularly radiostrontium, in the presence of suspended solids are reported.

From the combined results of the literature collation and of experimental data, design criteria for a small-scale water decontamination unit are summarized. Previous designs and their performances, where actual data are available, are reviewed.

Chapter II

RADIONUCLIDE REMOVAL BY ION EXCHANGE

The ion exchange process, first reported and systematically investigated over a hundred years ago, has made its most significant advances within the last fifteen or so years, and it is generally considered to have the status of a standard unit process now. Various theories have been proposed to account for the observed equilibrium and kinetic relationships between ionic species bound by the solid phase and those in the liquid phase, and much of the current knowledge of the ion exchange process has been summarized by Helfferich (14). Approximations of the more rigorous theory are available for working relationships and when radionuclides in trace concentrations are concerned, additional simplications are possible (15) (16) (17).

It is desirable to review elementary theoretical concepts applicable to radionuclide exchange, to indicate the nature and the significance of certain limitations of the process, and to review briefly previous investigations relating to decontamination.

Theoretical Considerations

The addition of ion exchange resin to a solution containing one or more ionic species initiates a series of reactions which redistribute the various ions between the resin and the solution, and these reactions continue until such time as no further change in the chemical composition of the solution occurs. Under this condition, the rate of sorption of each ionic species is equal to its rate of desorption and the overall effect is that of a static equilibrium situation. Among the simpler approaches to a formulation of the equilibrium state are those of the mass action principle, the Langmuir adsorption isotherm, and the Donnan membrane equilibrium; with appropriate assumptions, all three lead to approximately

the same working equation. It has been found that ion exchange occurs on an equivalent basis, that it is reversible, and that conventional stoichiometric equations can be written for the exchange reaction.

For illustration, competition between cation A with valence "a" and cation B with valence "b" for the anionic sites R in the resin matrix results in an equilibrium state represented by

$$bA^{+a} + B_aR \rightleftharpoons aB^{+b} + A_bR \tag{2}$$

and an analogous expression may be written for anion exchange. Application of the mass action concept for an equilibrium expression implies that equation (2) is equivalent to the usual metathesis reaction, and the restraining nature of the resin matrix is neglected. If it is further assumed that activity coefficients of the ionic species may be neglected in both the liquid and solid phases, a simplified equilibrium expression may be written as

$$K = \frac{c_B^a \; q_A^b}{c_A^b \; q_B^a} \tag{3}$$

where K is the selectivity coefficient, c is the concentration in the solution, q is the concentration in the resin phase, and the subscripts A and B refer to the respective ionic species.

In the special case of symmetrical exchange (a=b) between radiocation A of trace concentration and a single cationic species B of gross chemical concentration, virtually all the exchange capacity of the resin will be utilized by cation B, i.e. $q_A + q_B = Q$ and $q_B = Q$, where Q is the total exchange capacity of the resin. For this case, modification of equation (3) leads to

$$K = \frac{c_B \; q_A}{c_A \; Q} \tag{4}$$

which indicates that the distribution of radiocation A between the resin and liquid phases (q/c) is dictated by the value of the selectivity coefficient K, the concentration of B in the solution, and the total exchange capacity of the resin. For example, if K remains essentially constant and the concentration of B in the solution increases, less of the radiocation A will be sorbed by the resin at chemical equilibrium.

If a volume V' of solution containing given concentrations of gross cation B and of trace radiocation A is brought into contact with a given amount M of resin in the B form, the equilibrium distribution of A between the resin and liquid phases will approximate that predicted by equation (4). If this solution is withdrawn after equilibrium has been achieved, some of the radiocation A will remain with the resin phase. Introduction of another volume V' of the same initial solution would permit additional uptake of A by the resin, but the amount sorbed from the second batch would be less than that from the first. If this process is continued for n identical batches, where each batch achieves equilibrium before withdrawal, it may be shown that

$$F_n = \frac{n - \sum_{1}^{n-1} F}{K \frac{Q}{c_B} \frac{M}{V'} + 1} \tag{5}$$

where F represents the fractional attainment of the maximum uptake of radiocation A by the resin at equilibrium with the influent solution.

The preceeding material has considered batch operation; the theory for column operation where a solution is continuously passed through a fixed bed of exchange media is considerably more complex. Complicating factors, in addition to hydraulic dispersion effects, include the rate of diffusion through the liquid film surrounding the resin particles, the rate of diffusion within the particles, and the rate of reaction at the exchange site. The theories are presented an

discussed in Helfferich (14), and semi-empiric methods for data correlation and extrapolation have been proposed by Hiester and Vermeulen (18) and by Glueckauf (19).

The breakthrough curve for an ionic species passing through a resin bed initially in another chemical form is represented by a plot of the effluent concentration of the given influent ionic species against the solution volume passed through the bed, and in general the breakthrough curve is of an approximate sigmoidal shape. When the chemical compositions of the influent and effluent solutions are identical, the resin is at equilibrium with the influent solution. The amount of the influent ionic species retained by the resin bed may be determined from the breakthrough curve; the volume of solution with the given influent concentration which is equivalent to the total amount of the ionic species retained by the bed is approximately equal to the average breakthrough volume $\overline{V}$ for that ionic species.

In the case of an influent solution containing trace concentrations of radiocation A and gross concentrations of cation B, it may be shown (16) that the average breakthrough volume for the radiocation A is approximately

$$\overline{V}_A = K \ \overline{V}_B = K \ \frac{MQ}{C_B} \tag{6}$$

for symmetrical exchange. Since the average breakthrough volume of the gross cation B equals the total exchange capacity of the resin bed (weight of resin multiplied by the exchange capacity per unit weight) divided by the influent concentration of B, the average breakthrough volume of A is inversely related to the influent concentration of B.

The performances of the batch method and of the column method are illustrated in Figure 2 for the removal of radiostrontium in the presence of calcium. For this example of symmetrical exchange the following values have been assumed; $K = 1.4$,

Assumed Conditions:

$K_{Sr-Ca} = 1.4$
$Q = 5$ meq/g
$M = 20$ g.
$C_{Ca} = 3.6$ meq/liter

Column Operation

Batch Operation

Volume of Treated Water, Liters

Radiostrontium Concentration in Effluent, C/C_0
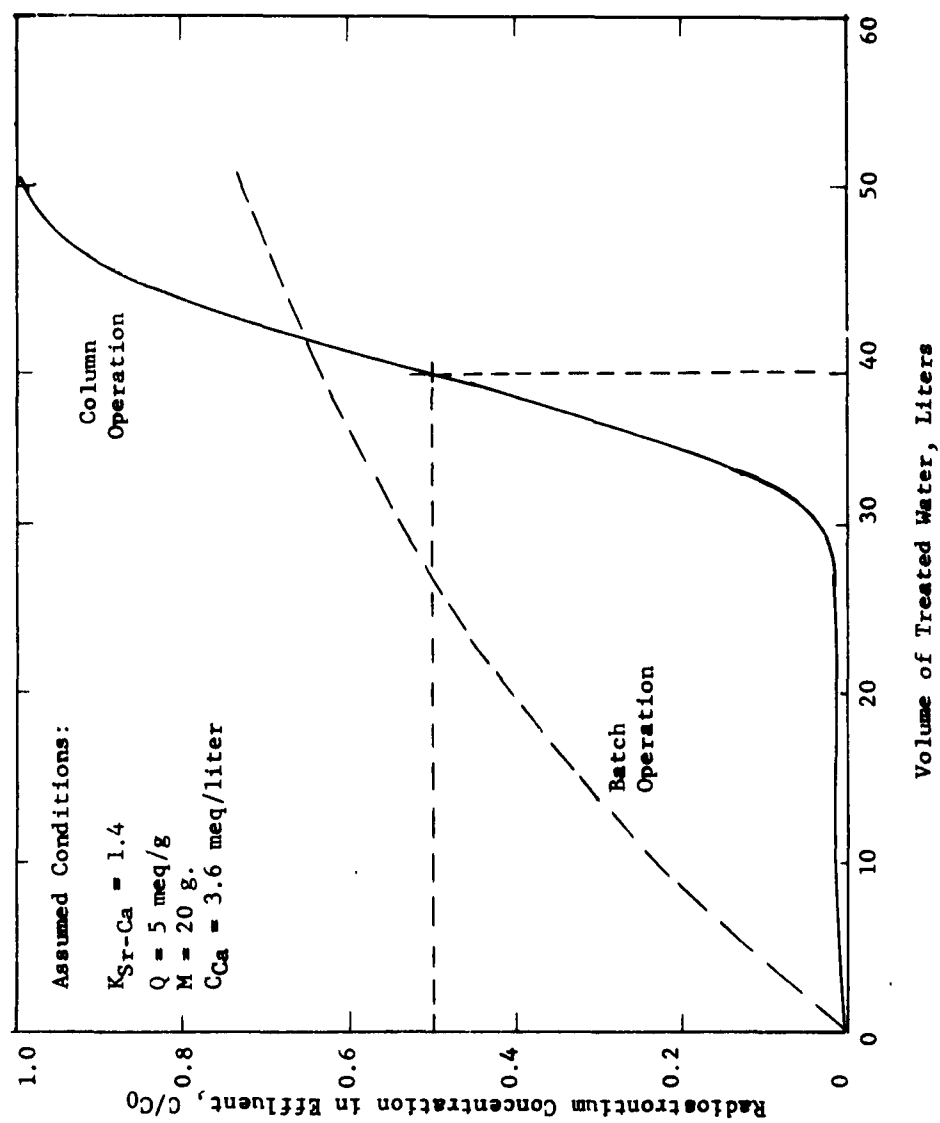
FIGURE 2. ILLUSTRATION OF ION EXCHANGE TECHNIQUES FOR RADIONUCLIDE REMOVAL

Q = 5 meq/g, M = 20 g, and the influent calcium concentration $(c_{Ca})_o$ = 0.0036 meq/ml. The dashed line representing batch operation is the limiting condition as the assumed V' magnitude approaches zero.

Although this discussion has used symmetrical cation exchange as an example because of its relatively simple formulation, analogous relationships may be derived for non-symmetrical exchange systems and for anion exchange.

Limitations of Ion Exchange

Ion exchange materials are not able to remove radionuclides from water after a state of chemical equilibrium has been reached, and complete removal is rarely, if ever, encountered. There is a two-fold objective in adapting the ion exchange process to water decontamination; to remove the radiocontaminants from as much water as possible and to provide a high degree of decontamination while doing so, but these objectives are not necessarily compatible. The limitations of the ion exchange process result in less effective decontamination for smaller volumes of water, and it is desirable to review the various aspects associated with these limitations.

It is assumed that column operation is superior to batch operation in water decontamination. Although both methods are capable of removing the same total amount of radionuclides, the column process, as illustrated in Figure 2, is capable of producing more water with greater decontamination. The present discussion is restricted to column operation.

The volume of decontaminated water produced by an ion exchange column is indicated approximately by the average breakthrough volumes for the individual radionuclides, but these average breakthrough volumes are dictated by the gross chemical concentration (in ionic strength) of the influent waters, the value or

values of K, and by the total exchange capacity of the bed. The greater the gross chemical concentration of the water applied to an exchange bed, the smaller will be the volume of decontaminated water produced.

TABLE II

STRONTIUM-CALCIUM SELECTIVITY COEFFICIENT, K

Conditions	Value of K*	Reference
Values increase with higher degrees of cross-linkage; synthetic organic cation resin	1.13 1.26 1.39	(20)
Values decrease with more dilute solutions; synthetic organic cation resin.	1.29 1.12 others**	(21)
Observed variation with different media and measurement techniques; natural sands and clays.	1.13 to 1.61	(15)
Values decrease with more dilute solutions; synthetic organic cation resin. Batch method.	1.55 to 1.20 others**	(17)
Observed variation with different experimental conditions; synthetic organic cation resin. Column method.	2.00 to 1.38	(17)

* Based on equation (4)

** Values were less than unity with dilute solutions; the lowest value reported was 0.020. The results were obtained through the use of batch techniques, and it is not believed that these values are of significance in practical column operation.

There is also a certain amount of approximation involved when a value of K is assigned to a particular situation. For example, values of K applicable to the radiostrontium-calcium system of exchange are drawn from various references and reported in Table II. Several of these values were calculated from reported data and the overall range shown is from 1.12 to 2.00; a common design value for this exchange system is 1.4 or 1.5. For non-symmetrical systems, such as radiocesium-calcium, it is expected that the range in value of K would be much greater.

From a number of typical breakthrough curves it appears that the beginning of the radionuclide breakthrough roughly coincides with the gross chemical break-through; this is believed to be sufficiently characteristic to serve as an indicator of the radionuclide breakthrough. Withdrawing the column from operation when the gross chemical breakthrough occurs means a deliberate sacrifice of the remaining capacity of the resin for additional removal of radionuclides, however it offers the advantage of simple techniques for indicating the limit of usefulness of the resin bed. Any one of several methods are available to indicate the gross chemical breakthrough, but no simple technique is presently available for the detection of the radionuclide breakthrough.

Provided the column operation is halted when gross chemical breakthrough occurs, the limitation to the degree of decontamination is provided by the amount of radionuclides appearing in the column effluent prior to the onset of the conventional breakthrough curve. This "leakage" or "premature breakthrough" is seen in Figure 2. Although it is conceivable that leakage might result from short-circuiting flow through the bed or from a rate-limiting diffusional step, most previous work has been based upon the concept of non-exchangeable or non-electrolyte forms of the radionuclides. The exchange media are available to remove electrolytes from the influent solution, and the mechanism of ion exchange will not be operative on non-electrolyte forms. Other mechanisms, such as surface adsorption through van der Waals' forces, might partially be successful, but this becomes, and should be treated as, a special case.

The low solubility of fallout materials indicates that much of the radiocontaminant present in the influent waters will be incorporated with colloidal size particulates, and it is to be expected that these particles would not be removed from the solution by an ion exchange mechanism. The soluble fraction of the fallout materials may become non-exchangeable, at least in part, through sorption by suspended solids or through the formation of radiocolloids.

The term 'radiocolloid' has been applied to those radionuclides in solution whose behavior is more suggestive of colloids than of simple ions. Most of the earlier experimental work on radiocolloids was performed by European investigators on the radioelements of the heavy metals which possess hydroxide forms of low solubility, but more recent work by Schubert (22) has indicated that, under appropriate conditions, any radionuclide could assume a colloidal form.

Theories which have been advanced to explain radiocolloid formation include (a) that of presumed sorption of the radionuclides by foreign impurities in the solution and (b) that of the formation of aggregates of colloidal size by hydrolyzed forms of the radionuclides. The validity of one theory over the other was a source of contention for many years, but the present concept holds that both mechanisms probably contribute to the formation of radiocolloids in a given case (17).

From the results of many early investigations it appears that polyvalent radiocations such as yttrium, lanthanum, zirconium, lead, bismuth and niobium are able to form hydrosols under proper pH conditions in either distilled or tap waters, but the amount of hydrolysis is restricted in acid waters. Radiocations of strontium, barium, cesium, and sodium apparently have little tendency to hydrolyze in natural waters. Since the polyvalent cations are but sparingly soluble from the fallout form, it would seem that radiocolloids resulting from hydrolysis are of little significance in the surface waters following a nuclear attack.

The sorption of mono- and divalent radiocations by the suspended solids present in waters probably will be of importance, since these forms will be unavailable for ion exchange because of non-electrolyte formation. Eisenbud (23) summarises much of the current data relating to the uptake of the fission products by the biological life forms associated with surface waters and a number of studies illustrating the sorption of fission products by clay minerals in water have been made (24) (25) ·

That colloid formation leads to a modified affinity of exchange media for radiocations can be illustrated by the work of Gustavson (26), Adams (27), Schubert (28), Holm (29), Sussman et al. (30) and Bunney et al. (31); these studies have been directed toward the understanding of the differing chemical states of an element in solution and have employed ion exchange as a separation technique.

By way of general summary, it may be said that the volume of water which can be decontaminated is primarily dictated by the chemical concentration or ionic strength of the natural water, and that the limitation of decontamination effectiveness is determined by the presence of non-exchangeable or non-electrolyte forms of the radionuclides.

Previous Investigations

Utilization of the ion exchange process for the treatment of radioactive laboratory wastes was studied by Ayres (32); he envisioned the leakage of radionuclides as resulting from their extremely small chemical concentrations and suggested that the addition of carrier ions to achieve greater removal by the process of mass action. This suggestion would lead to early saturation of the exchange media and would reduce the total volume of water decontaminated by a given column.

Lacy and Linsten (33) investigated the removal of mixed fission products from tap water and reported greater decontamination effectiveness at lower flow rates. Swope has investigated both cation (34) and mixed resin (35) beds for the removal of mixed fission products with attention being given to chemical form of the resin, the influent pH, the flow rate, and the resin cross-linkage. The mixed bed was reported as being more effective in decontamination degree than the cation bed, but the flow rate did not appear to be significant within a range of 2 to 10 gpm/sq ft. Hydrogen form resin appeared to be superior to the other forms of resin in the removal of alpha emitters, but significant differences in removal of beta emitters were not found. Greater decontamination was demonstrated at lower pH values of the influent solutions and by passing the solution through a second resin column in series.

Decontamination effectiveness in the removal of iodine-131 and of strontium-89 with cation resin, anion resin and mixed resin was reported by Friend (36). Although demonstrating removals in excess of 99%, certain irregularities in the radioidine leakage zone were attributed to different chemical forms, and the radiostrontium removals were probably greater than reported because of radioyttrium leakage.

The effects of flow rate, column length, resin particle size, temperature and pH of the influent solution on radiostrontium leakage were investigated by Lacy (37) using a high capacity strong acid cation exchanger. Although small in magnitude, increased removals were obtained with lower flow rates, increased column lengths, smaller resin particle size, higher temperature and increased pH.

Klein et al. (38) studied the leakages of yttrium-91 and of barium-lanthanum-140 through natural greensand exchange columns and through synthetic resin columns. Both yttrium and lanthanum exhibited radiocolloid properties, particularly at higher pH values, and their removals decreased accordingly. Mixed resin beds were

found superior to cation resin beds. The degree of removal varied inversely with flow rate.

Other than Klein's studies, the variation of the chemical state of radioyttrium in aqueous solution has been investigated by Bain (39) using batch exchange studies, Kurbatov and Kurbatov (40) using filter paper, and Ray (17) using membrane filter techniques. It appears that radioyttrium exists primarily in the cation form at pH values below 4 or 5 and that an abrupt change toward non-ionic or anionic forms occurs in the pH range from 6 to 7.

Earlier studies (16) (17) were concerned with the leakage of radiostrontium a 24 - in deep bed of cation resin. Based on an assumed sorption of a fraction of the total radiostrontium in the influent solution by the suspended solids, the significant leakage parameters were indicated as the initial amount sorbed by the suspended solids, the effective residence time of the solution within the resin bed and an empirical rate constant of dissociation from the solids. Similar studies were made of the dissociation of radioyttrium; the pH of the solution was of importance because of its influence on the chemical state of radioyttrium.

Chapter III

CHARACTERISTICS OF SURFACE WATERS

Considering the several alternate sources of water following a large scale
nuclear event, those waters most likely to be contaminated significantly are those
subject to direct fallout such as lakes, rivers and smaller streams. In some cases,
waters in a distribution system, particularly if open reservoirs are present, could
also contain radiocontaminants. As a first approximation, ground water or well
water may be assumed to have little contamination, at least in the early emergency
phase.

Since the decontamination of water by ion exchange is related directly to
the characteristics of the water itself, it is necessary to estimate the range
in values for each important characteristic which might be reasonably anticipated.

Importance of Suspended Solids

Previous laboratory decontamination studies have been directed primarily
toward those in which reactor-produced radionuclides have been added to synthetic
waters in an attempt to simulate probable field conditions.   Most of these
studies have omitted suspended solids, presumably because of an implied pre-
treatment process; in the situation assumed to follow a nuclear event such
allowance does not appear to be justified.

Sample calculations may be used to illustrate the significance of the
presence of suspended solids in the influent waters. If the water contains about
70 mg/l (weight basis) turbidity with an exchange capacity of 50 meq/100 g, and
if the predominate cation is calcium at a concentration of 100 ppm $CaCO_3$ (0.002N),
the turbidity would incorporate about 2.6% of the total radiostrontium. This
calculated value, based on equation (4), assumes a selectivity coefficient of
1.5 for the strontium-calcium system.

A similar calculation with assumptions of 50 ppm CaCO$_3$ (0 001N) calcium and
of 500 mg/1 turbidity leads to an estimate of over 25% of the radiostrontium in
the solid phase.

The decontamination factor for radiostrontium would range from 50 to 4 for
the cited conditions if the turbidity passed through the resin bed without loss
and if the radiostrontium intially sorbed by the turbidity did not dissociate
during passage through the resin. These figures, which are not unnecessarily
extreme, tend to refute a casual assumption that ion exchange is capable of
providing decontamination factors of 1,000 to 10,000 for radiostrontium.

Since the calculations were based upon the soluble fraction of the total
radiostrontium, it might be anticipated that passage of the insoluble portion,
incorporated by the fallout material, could decrease the calculated decontamina-
tion factor even more.

Variations in Turbidity and Total Hardness

The indicated calculations of the preceeding section were based upon
assumed values of turbidity and of total hardness, and it is desirable to
determine how realistic such values might be in the surface waters of the
continental United States. Because of the known variations with time of turbidity
and of total hardness at any one location and of rather large variations with
geographic location, it is obvious that considerable error is likely without an
extensive study of appropriate data.

Perhaps the most meaningful grouping of turbidity and total hardness data
would reflect values to be anticipated in the waters likely to receive significant
levels of radiological contamination and subject to consumption without pretreatment.
Some allowance should be made in terms of the volumes of water and the consumer
populations; weighed average values conceivably are possible both for turbidity
and for total hardness.

Since an indication of the variations is sufficient for illustration in this report, and since the compilation of required data is a tangential effort, simplifying assumptions were arbitrarily made. Data reported by Nordell (41) and by the United States Geological Survey (42) were abstracted and tabulated: average values of turbidity and of total hardness were assigned to each state on the basis of one value per five million population, and these values were numerically ranked in order.

Probability plots of total hardness and of turbidity are shown in Figures 3 and 4, respectively. These data plots suggest that a median value of total hardness is about 130 mg/l as calcium carbonate, and a similar median value for turbidity is approximately 70 ppm. Ninety percent of all total hardness values lie between 20 and 530 mg/l as calcium carbonate, and ninety percent of the turbidity values are from less than 10 to about 800 ppm.

While the absolute validity of these values is questionable, there is little doubt but that considerable variations both in turbidity and in total hardness can reasonably be anticipated in the surface waters of the continental United States following a nuclear attack.

The necessity of providing a means for detecting resin exhaustion may be illustrated from these data. Assuming the cation exchange resin in a decontamination to possess a total exchange capacity of 650 meq (for example, 130 g of resin with an exchange capacity of 5 meq/g) and the influent water of 130 mg/l total hardness as calcium carbonate, the unit would be able to process almost 250 liters before exhaustion. However, if the water hardness is 530 mg/l the treated volume decreases to 61 liters, but if the water hardness is only 20 mg/l a total of about 1,620 liters can be processed. The only way to insure that the resin will be fully utilized while giving reasonable assurance of decontamination is to provide a suitable means for breakthrough or exhaustion indication.

Indicated values derived from data reported in references (41) and (42).

Percent Equal to or Less than Indicated Value

Total Hardness, mg./liter as CaCO₃
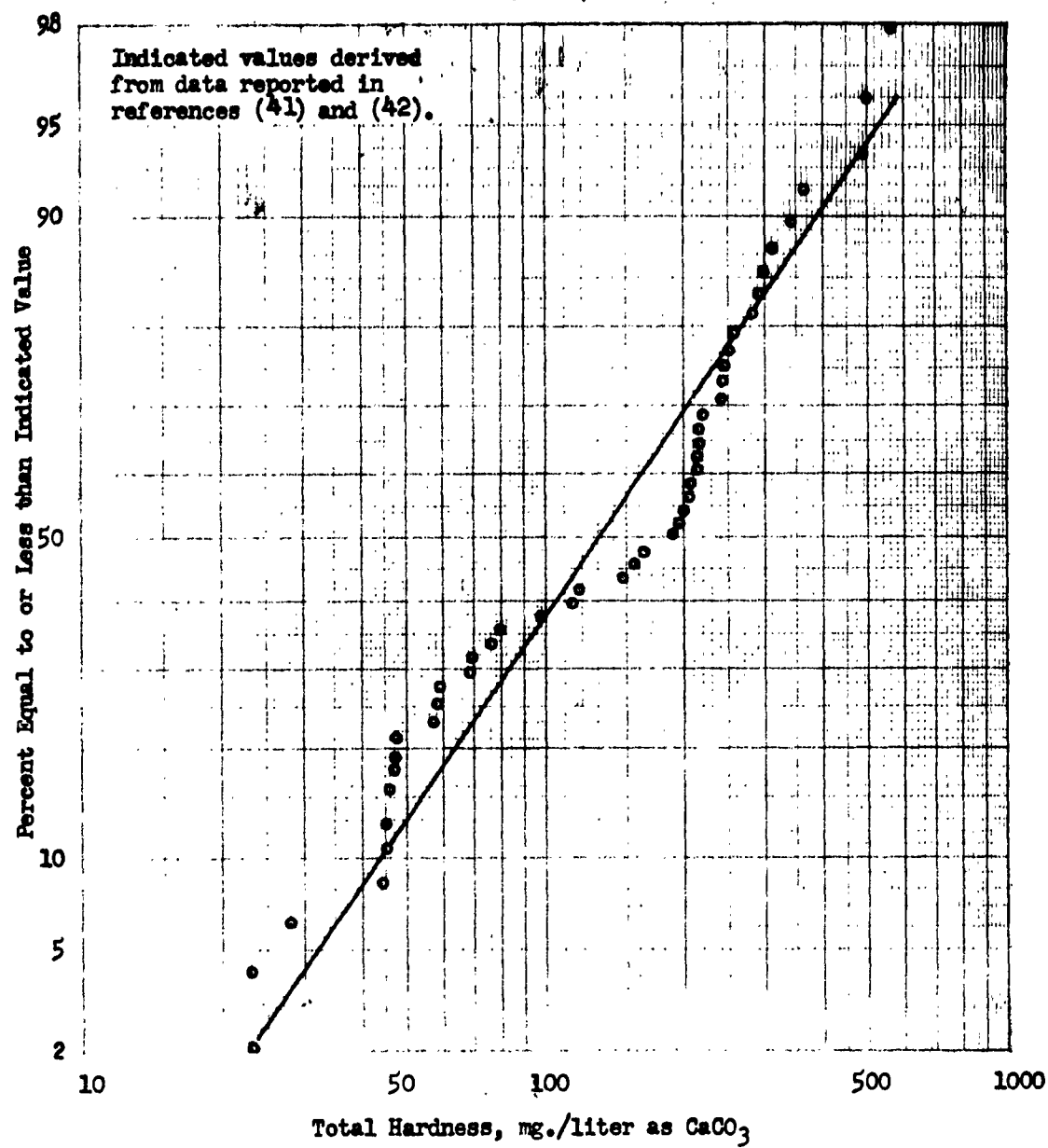
FIGURE 3. GENERAL VARIATION OF TOTAL HARDNESS IN SURFACE WATERS
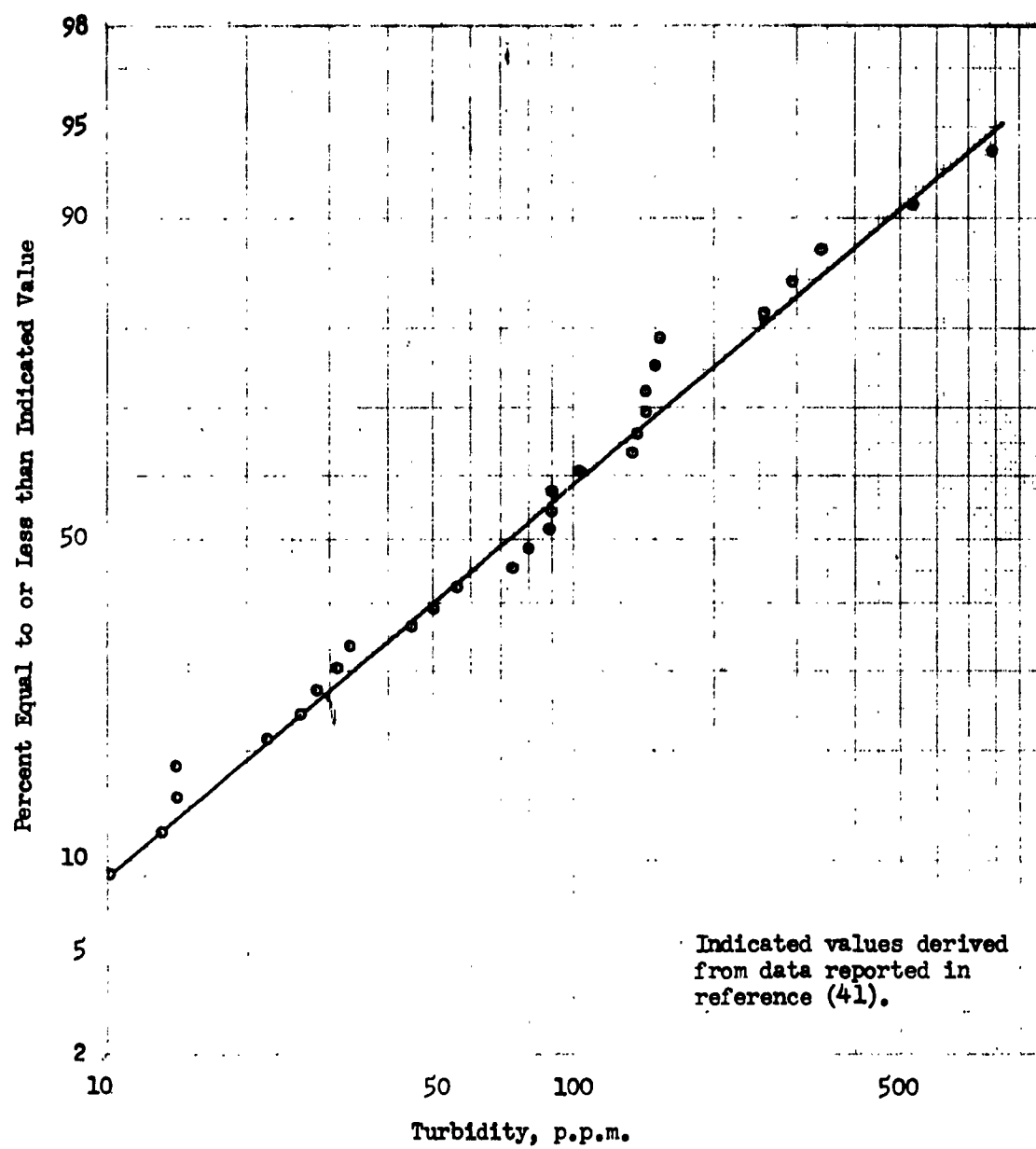
FIGURE 4. GENERAL VARIATION OF TURBIDITY IN SURFACE WATERS

Chapter IV

FILTRATION THEORY

In order to obtain some perspective on the interaction of raw water turbidity with ion exchange resins in the absence of pretreatment, reference is made to the literature dealing with the conventional rapid sand filtration process.

The application of raw surface waters to beds of ion exchange resins is somewhat analogous to the rapid sand filtration process commonly found in water treatment plants. Although considerable differences are to be found between charged porous solid resin beads and neutral non-porous solid sand particles, from a hydrodynamic view they both represent beds of granular solids through which water passes. The many different organic and inorganic materials making up the turbidity of raw waters is replaced by the flocculent suspensions carried over from the sedimentation units in conventional water treatment plants, but in a gross sense they are all suspended solids which conceivably are subject to removal, at least in part, by the filtration process.

Despite the many studies which have been conducted on the filtration process in beds of granular solids, the nature and the relative importance of different removal mechanisms have yet to be determined. At this time it appears that a simple expression for filtration effectiveness is unlikely except as an acknowledged approximation of a more exact expression; moreover an exact expression has yet to be formulated and experimentally demonstrated as valid over the entire range of possible filtering conditions.

A great deal of practical information and operating experience with the sand filtration of water supplies has been contributed by Baylis (43), and a committee of the American Society of Civil Engineers (44) made one of the first major efforts to identify some of the basic problems involved in rapid sand filtration.

During the last thirty years a number of significant contributions, among which are those by Eliasson (45), Stein (46), Iwasaki (47) Hudson (48), Stanley (49), Hall (50), Ghosh (51), Ives (52) and Mackle and Mackle (53), have been made toward a better understanding of the problems involved. It is not considered to be within the scope of this report to review in detail the work of each individual: it appears that a summary of the current status of knowledge and concepts dealing with the removal of suspended solids by granular solids beds is more desirable.

Most of the experimental work done previously has been accomplished with influent waters containing flocculated suspensions prepared with various coagulants, Notable exceptions have been the Fullers earth suspensions employed by Ghosh (51) and the radioactive algae used by Ives (52).

Water filtration on a plant scale may be limited either by the appearance of excessive amounts of turbidity in the filtrate or by the creation of excessive head loss through the bed. One approach to an adequate basis of design is typified by an equation of the form

$$K_f = \frac{d^l h^m Q^n}{L} \tag{7}$$

where d is the effective size of filter medium grains, L the bed depth, h the terminal head loss, and Q the application rate on the bed influent. The coefficient K_f is presumably an index to filtration effectiveness in the process involving given suspended solids and a certain filter bed, and its value depends upon the plant performance observed over a period of time. The exponents l, m, and n have been assigned values of 3, 1, and 1 and of 2.46, 1, and 1.56 by Hudson (48) and by Stanley (49) respectively. Application of this equation is to be found in compensation for a change in one variable so the overall filtration effectiveness is not impaired. For example, if the K_f value associated with termination of a filter run is known from observations made under operating conditions and it is desired to increase the operating flow rate, quantitative estimates for the

increased bed depth, the decreased grain diameter, or the smaller head loss
which will preserve the present filtration effectiveness may be obtained.

Attempts have been made in more recent years to formulate the kinetics of
the filtration process and to single out the more important removal mechanisms.
Stanley (49) has listed and briefly discussed possible contributions by straining,
sedimentation, inertia, contact action, electrokinetic force, Brownian movement,
and van der Waals forces to the filtration process. Hall (50) has attempted to
develop useable expressions for interstitial straining and gravitational sedi-
mentation mechanisms, and consideration of the van der Waals and hydrodynamic
forces has been made by Mackrle and Mackrle (53). It appears possible that the
primary removal mechanism is that of graviational sedimentation, aided to some
extent by interstititial straining, but probably adsorptive forces become dominate
when a particle approaches a grain surface.

The current concept of the filtration process, at least to its application
in beds of granular solids, is that the depth to which a solid particle will
penetrate is a result of two phenomena By one or more of the mechanisms
previously mentioned the particle is removed from the flowing stream and re-
tained by or near the solid grains. Superimposed upon the removal action is a
"creep flow" which is brought about by increased interstitial velocities
accompanying reductions in pore size.

Both Hall (50) and Ives (52) have assumed that the removal of suspended
material is proportional to its concentration in the water; this may be expressed
as

$$\frac{dc}{dz} = - r c \tag{8}$$

where c is the suspended material concentration in the water phase, z is the dis-
tance from the filter surface, and r is a general rate factor which is dependent
upon the particular mechanism or mechanisms involved in the filtering action.

Assuming a homogeneous isotropic porous medium, the separation mechanism the same throughout the filter, and independent behavior on the part of individual suspended particles (conditions are which most nearly met at the beginning of a run through a clean filter) Hall (50) has formulated expressions for the rate factors as either

$$r = \frac{a}{Q} \qquad \text{for gravitational sedimentation} \qquad (9)$$

or

$$r = \frac{b}{d^{3/2}} \qquad \text{for interstitial straining} \qquad (10)$$

In these expressions, a and b are constants, Q is rate of flow, and d is the grain size of the filter medium.

If gravitational sedimentation is assumed to be the primary mechanism, combining equations (8) and (9) leads to

$$\frac{c}{c_o} = e^{\frac{-a\ L}{Q}} \qquad (11)$$

where L is the bed depth, Q is the rate of flow, a is a constant, and c/c_o is the ratio of effluent concentration to the influent concentration for the suspended particles. Although this simplified expression is but an approximation for a quite complex phenomenon, the indicated trend of decreased effluent turbidity concentration with decreased flow rate and with increased bed depth has been experimentally demonstrated.

Ives (52), making allowance for a changing rate factor with solids deposition within the filter pores, has developed a more complete equation for the fil ering process which is not readily solved analytically, but solutions have been obtained with a digital computer using empirically determined constants.

From the results of a study on the removal of Fullers earth turbidity by either sand or by glass bead filter media, Ghosh (51) concluded that the turbidity penetrated the entire depth of the media under almost all conditions of flow, and

there was a gradual reduction in turbidity all along the depth. He also observed that the effluent turbidity, and hence the filtration effectiveness, depended more on the velocity of flow than on the size of the filter medium, and the initial concentration of the turbidity had little effect on the percent removal.

Chapter V

LABORATORY INVESTIGATIONS

The need for information on the relationship between suspended solids in waters to be treated and ion exchange resin beds led to appropriate laboratory studies; additional data on the interactions between suspended solids, radionuclides, and beds of exchange resins was also obtained.

The suspended solids employed in this investigation were from either local soils or reference clay minerals, the exchange media included various organic synthetic resins, and radiostrontium was chosen as representative of the radionuclide of health significance.

Leakage of Solids through Resin Beds

Stock solutions were prepared by adding those crushed solids materials passing a No. 200 mesh sieve to waters of the desired chemical composition. After an initial agitation, the solutions were allowed to stand overnight, and the supernatant was removed for use as a stock solution. The concentration of solids in the stock solution was determined on a weight basis by passing samples through membrane filters and weighing when dry. Analyses were made in triplicate and control filters were used to compensate for the change in weight of the membrane filter during analysis. The probable error of the mean concentration was determined as about 1.5%.

It was found that simple measurements of the solids concentrations could be obtained with a colorimeter* provided calibrations were made with membrane filters. The inherent problems of applying a light scattering technique toward a weight measurement were apparently minimal during a particular run, although the calibration curves differed between runs, and continuous checks on the calibrations were made with membrane filter weighings. A typical calibration curve, indicating conformance with the Beer-Lambert concept, is shown in Figure 6.

*Bausch and Lomb "Spectronic 20."

Influent solutions were obtained by dilution of the known stock concentration to the desired concentration, and the influent solutions were passed by gravity through the resin beds. The influent solutions were agitated continuously during each run to maintain a constant solids concentration, and the rate of flow was manually controlled with an adjustable clamp on the effluent line. This flow rate control method appeared to provide constant flow rates within 5% except when excessive amounts of solids accumulated within the pore spaces of the bed.

An analysis, the average of a number of such analyses, of the tap waters used in the investigation is given in Table III, and the composition of the local soil sample is presented in Table IV. The reference clay minerals, obtained commercially, included:

Illite No. 35	Kaolinite No. 4	Montmorillonite No. 20
Fithian, Illinois	Oneal Pit	Husband Mine (Bentonite)
	Macon, Georgia	Polkville, Mississippi

The column was loaded with resin by partially filling the column with distilled water and dropping the resin, a little at a time, into the water. Combining this procedure with a continuous tapping on the side of the column apparently provided reasonably reproducible beds without air in the void spaces. After a sufficient amount of resin (4 in. depth, approximately 50 ml) had been added, distilled water was passed downward through the bed. While the distilled water flow was provided primarily for flow rate regulation, it also improved the packing of the bed. A sketch of the column apparatus is shown as Figure 5.

Anion, cation, or mixed resin beds* were employed as desired for the various runs. The column effluent was fractionated into appropriate samples, and the solids concentration of each sample was determined with colorimeter.

Experimental data obtained with local soil suspended solids is given in Appendix A; that for the reference or standard clays in Appendix B. Plots of the data are normalized by referencing the effluent solids concentration and the throughput

*"Amberlite" resins IRA-400, IR-120, and MB-3; manufactured by Rohm and Haas, Philadelphia

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Table III

TAP WATER ANALYSIS*

Cations	(mg/1)
Sodium	4.1
Potassium	1.4
Calcium	27.1
Magnesium	0.6
Iron	0.05
Aluminum	0.26

Anions	(mg/1)
Chloride	6.5
Fluoride	1.0
Sulfate	35.
Alkalinity (as $CaCO_3$)	62.

* pH 8.4

Table IV

LOCAL SOIL SAMPLE COMPOSITION

| Component	Chemical Formula	Approximate Percent
Quartz	SiO_2	64
Kaolinite	$Al_2Si_2O_5(OH)_4$	19
Apatite	$3\ Ca_3P_2O_8 \cdot CaF_2$	15
Illite	$KAl_3Si_3O_{10}(OH)_2$	1
Iron Oxide	Fe_2O_3	1
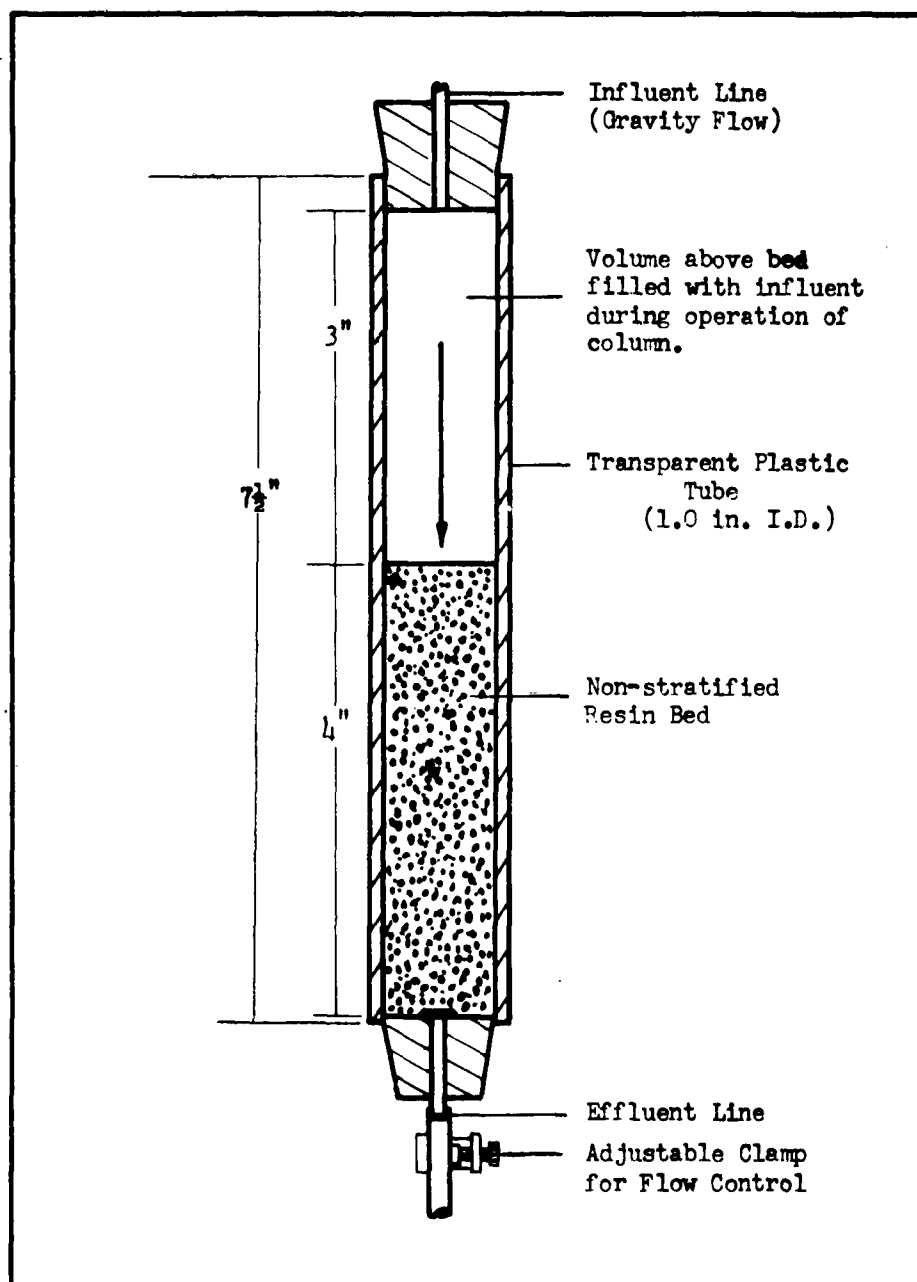
FIGURE 5.   SKETCH OF EXPERIMENTAL APPARATUS

volume to the influent concentration and the 50 ml bed volume, respectively, and these results are shown in Figures 7 through 13.

Under the conditions investigated, it was observed that suspended solids leakage occurred to some degree after the pore liquid had been displaced by the influent solution. Variations observed were in the amount and the nature of the leakage from anion resin beds in relation to ionic strength, nature of the suspended solids, and in the flow rate; under no circumstances was the resin successful in removing all of the suspended solids.

For example, a greater amount of the suspended solids was retained by the bed prior to the establishment of a constant rate of leakage for solutions of greater ionic strength. Virtually no solids retention was observed in cation resin beds, the leakage in anion resin beds was inversely related to the flow rate, and behavior of the leakage through mixed resin beds was intermediate between these extremes.

The nature of the retention forces was not clearly established, but certain observations indicated they were of small magnitude; for example, Figure 9 illustrates a self-flushing action encountered during a prolonged run in which the flow rate was maintained constant. Apparently the shearing stresses developed by the fluid were sufficiently large to cause a partial breaking up of the deposited solids in the bed.

Similar runs made with kaolinite and with illite clay minerals indicated behavior equivalent to that provided by the local soil solids, but bentonite clay did not penetrate the resin bed in significant amounts. This appeared to result from sedimentation on the surface of the bed and not from retention within the pore spaces.

The addition of a cotton filter pad above the resin bed was successful in removing the bulk of the suspended solids as shown in Figure 13, however some problem was encountered in eliminating air from the cotton pad and in maintaining a reasonable flow rate after the cotton had retained appreciable solids. A glass wool pad offered the same objections without provided the same degree of removal.
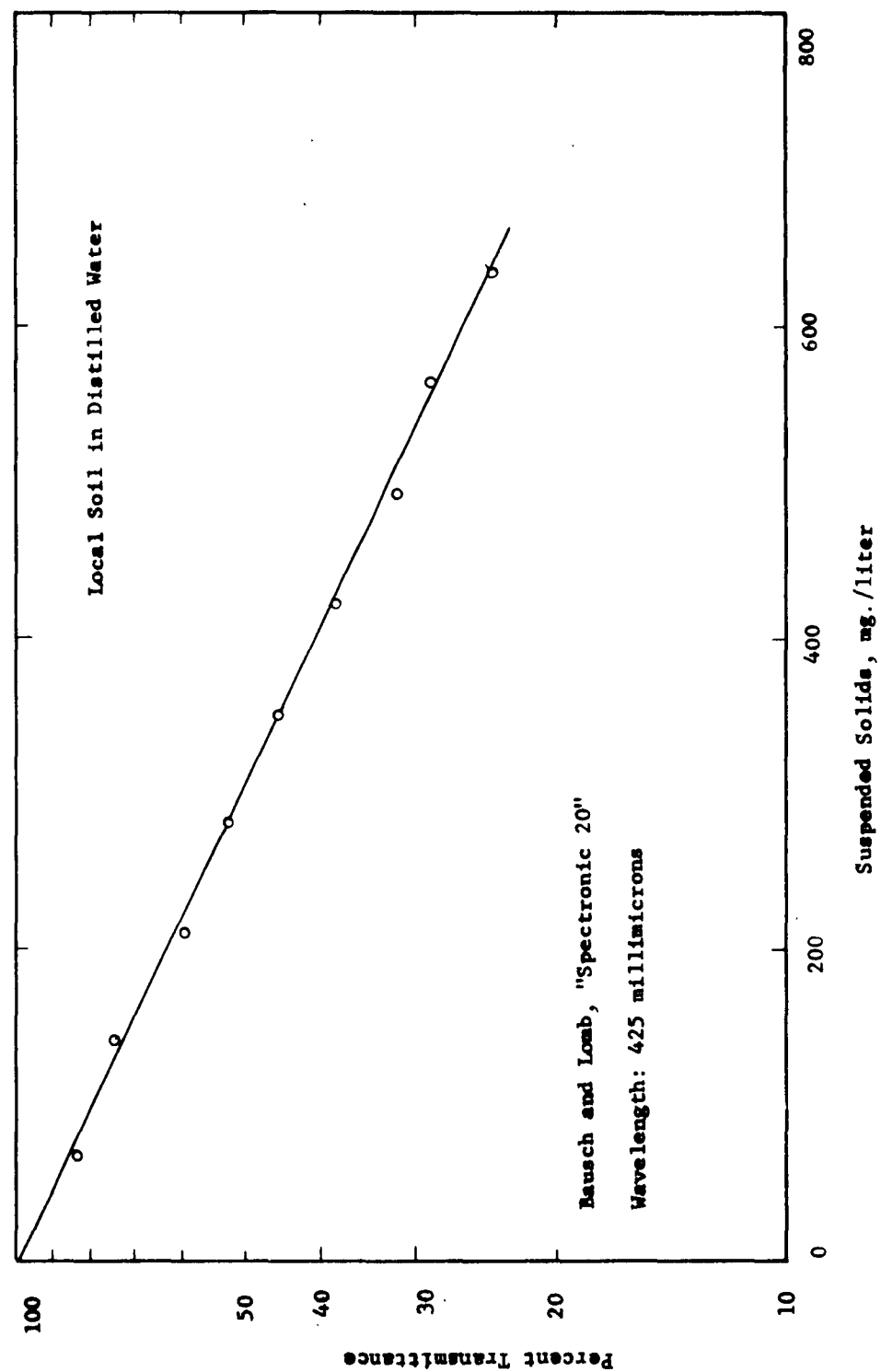
Figure 6. TYPICAL CALIBRATION CURVE FOR TURBIDITY MEASUREMENTS

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39
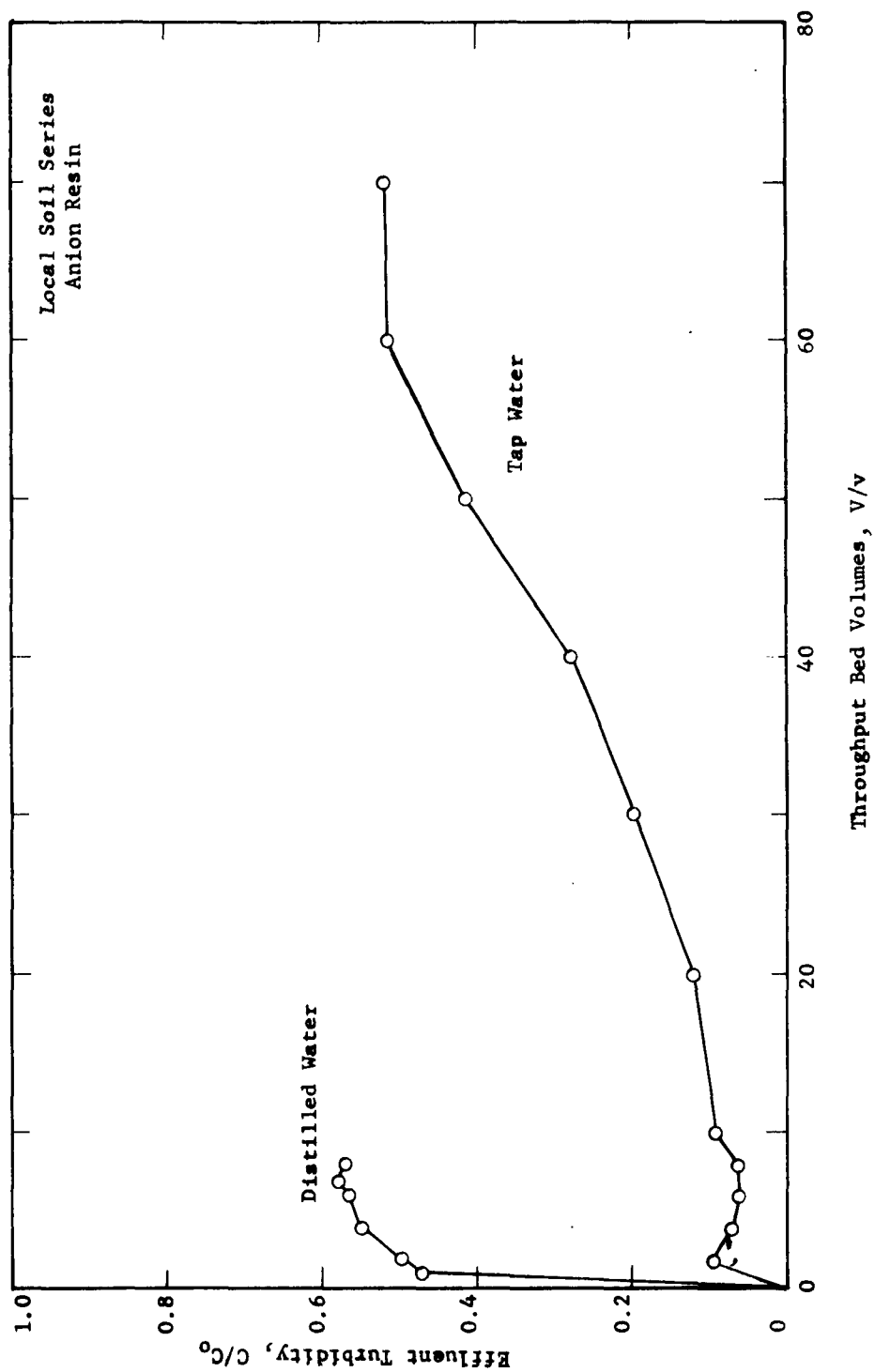
FIGURE 7. EFFECT OF IONIC STRENGTH ON SUSPENDED SOLIDS LEAKAGE
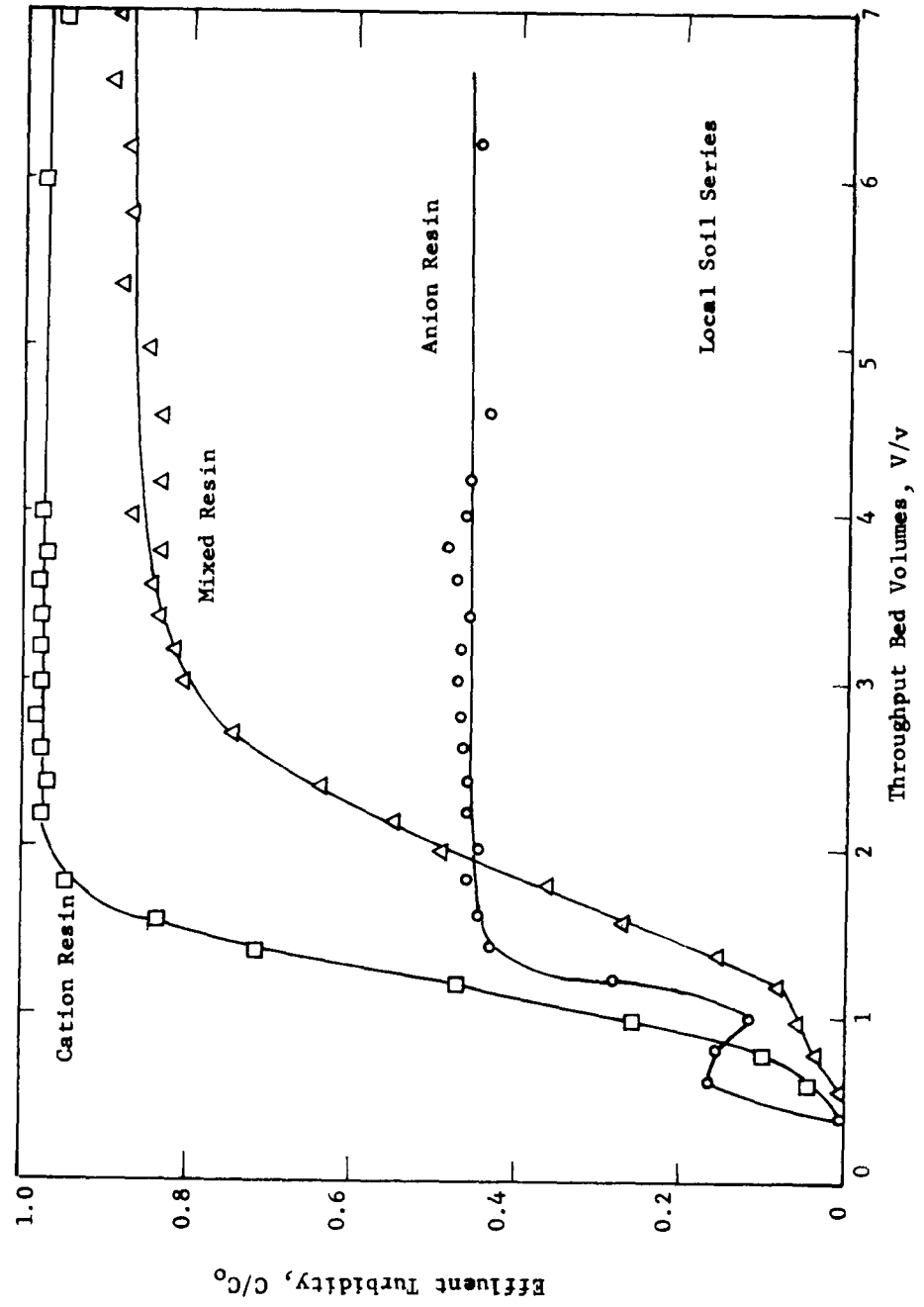
FIGURE 8. VARIATION OF SUSPENDED SOLIDS LEAKAGE WITH RESIN TYPE
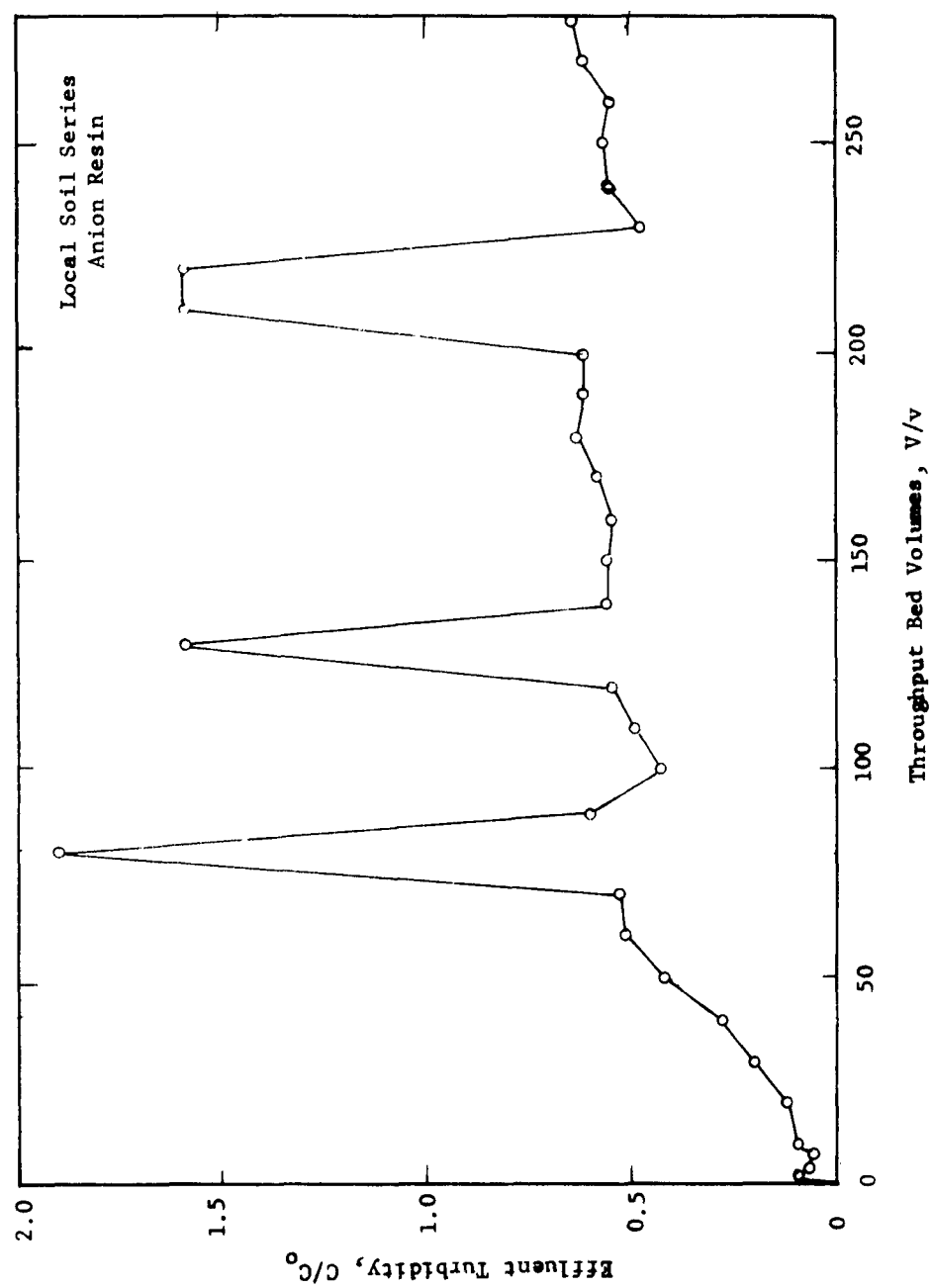
Figure 9: OBSERVED IRREGULARITIES IN SUSPENDED SOLIDS LEAKAGE
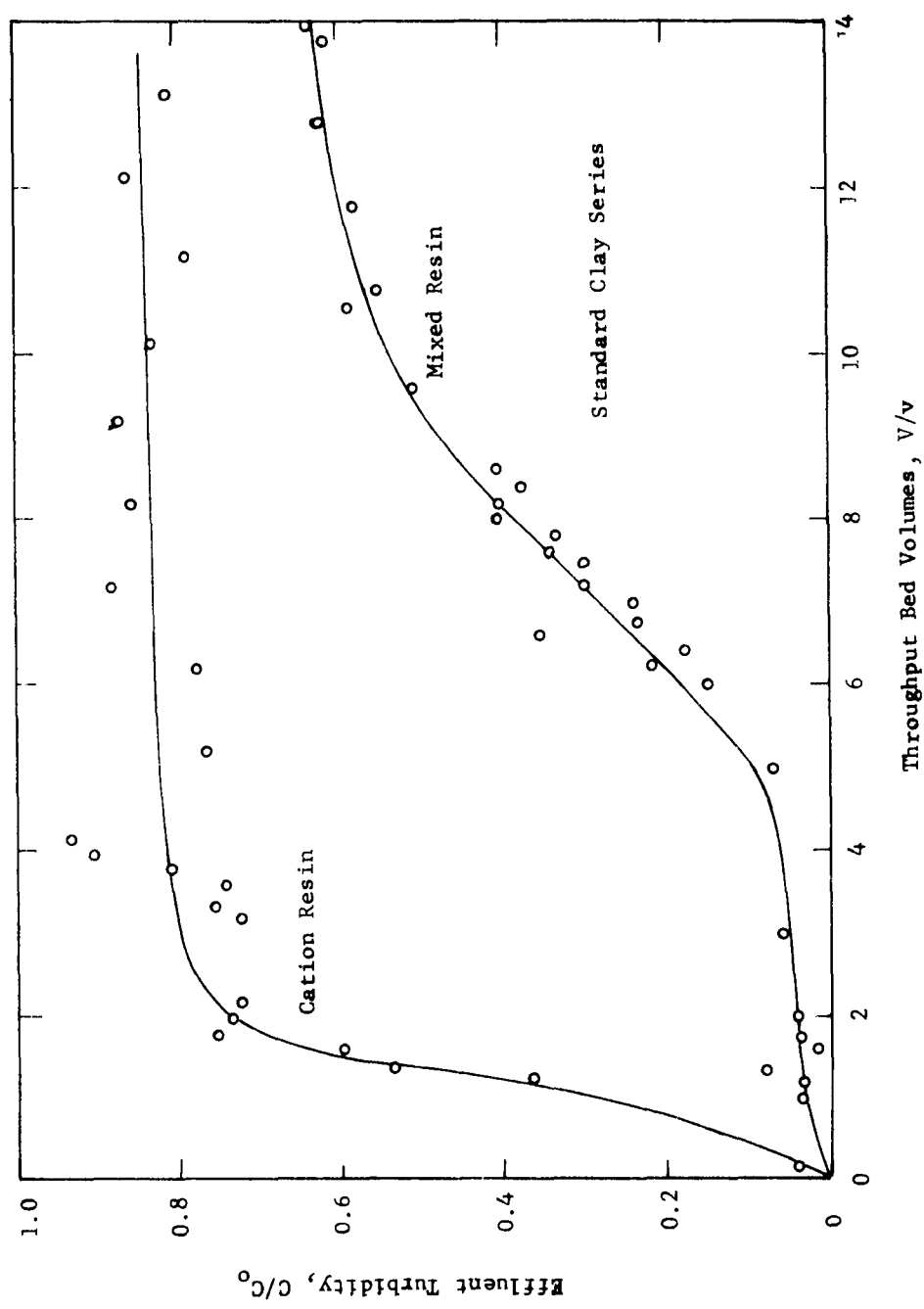
FIGURE 10. KAOLINITE LEAKAGE IN STANDARD CLAY SERIES

43
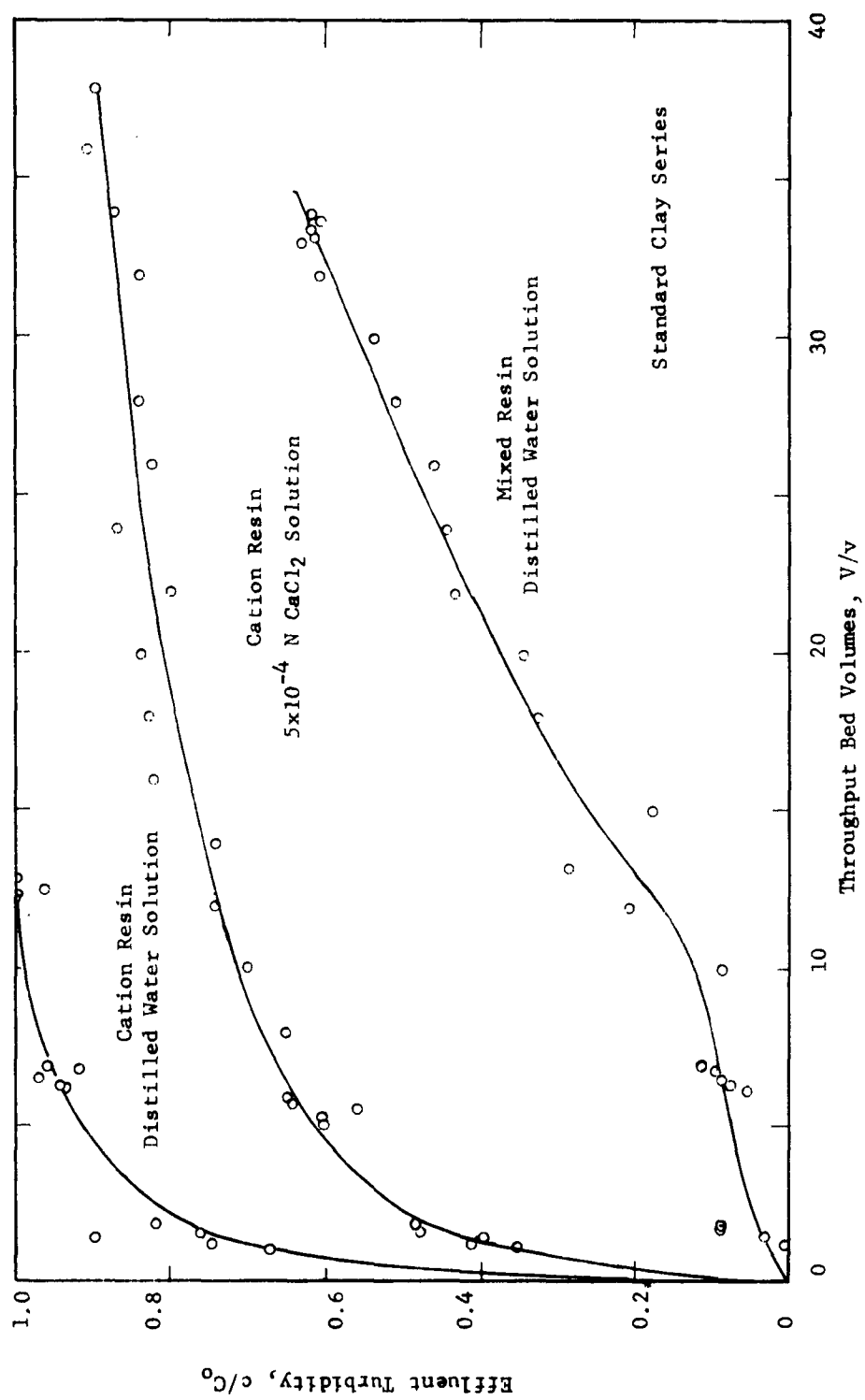
FIGURE 11. ILLITE LEAKAGE IN STANDARD CLAY SERIES

44
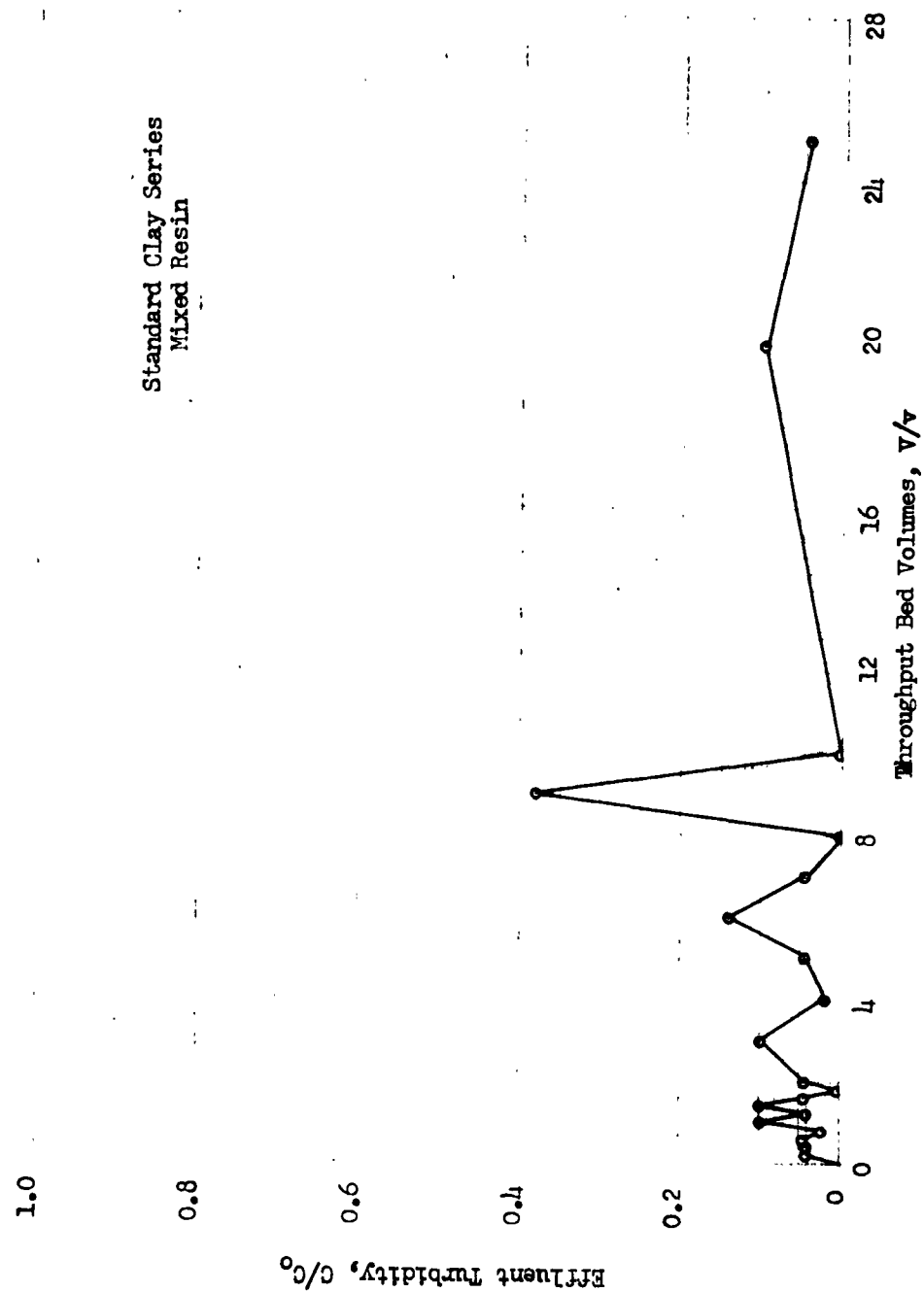
FIGURE 12. BENTONITE LEAKAGE IN STANDARD CLAY SERIES
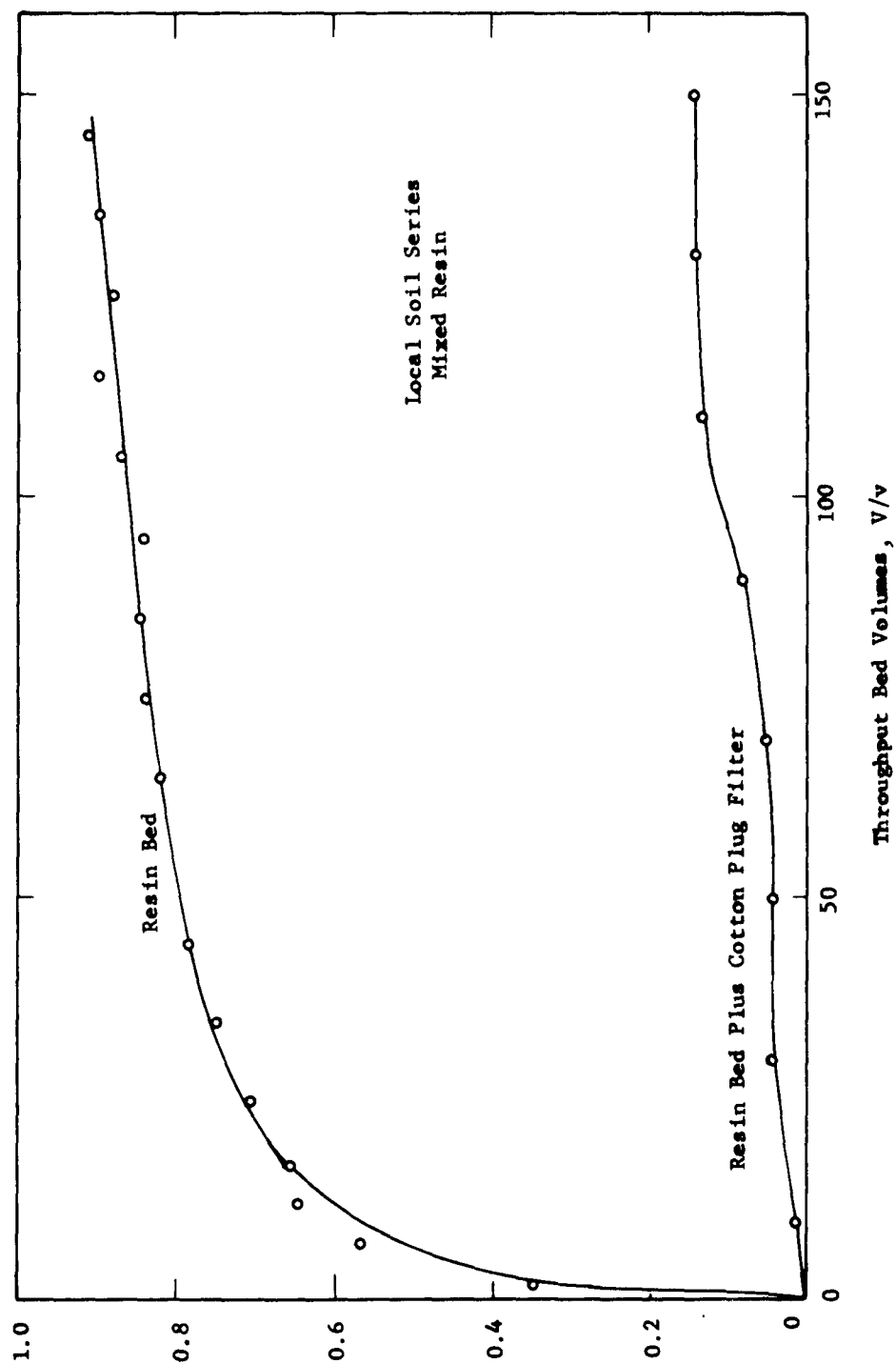
Figure 13. EFFECT OF FILTRATION ON SUSPENDED SOLIDS LEAKAGE
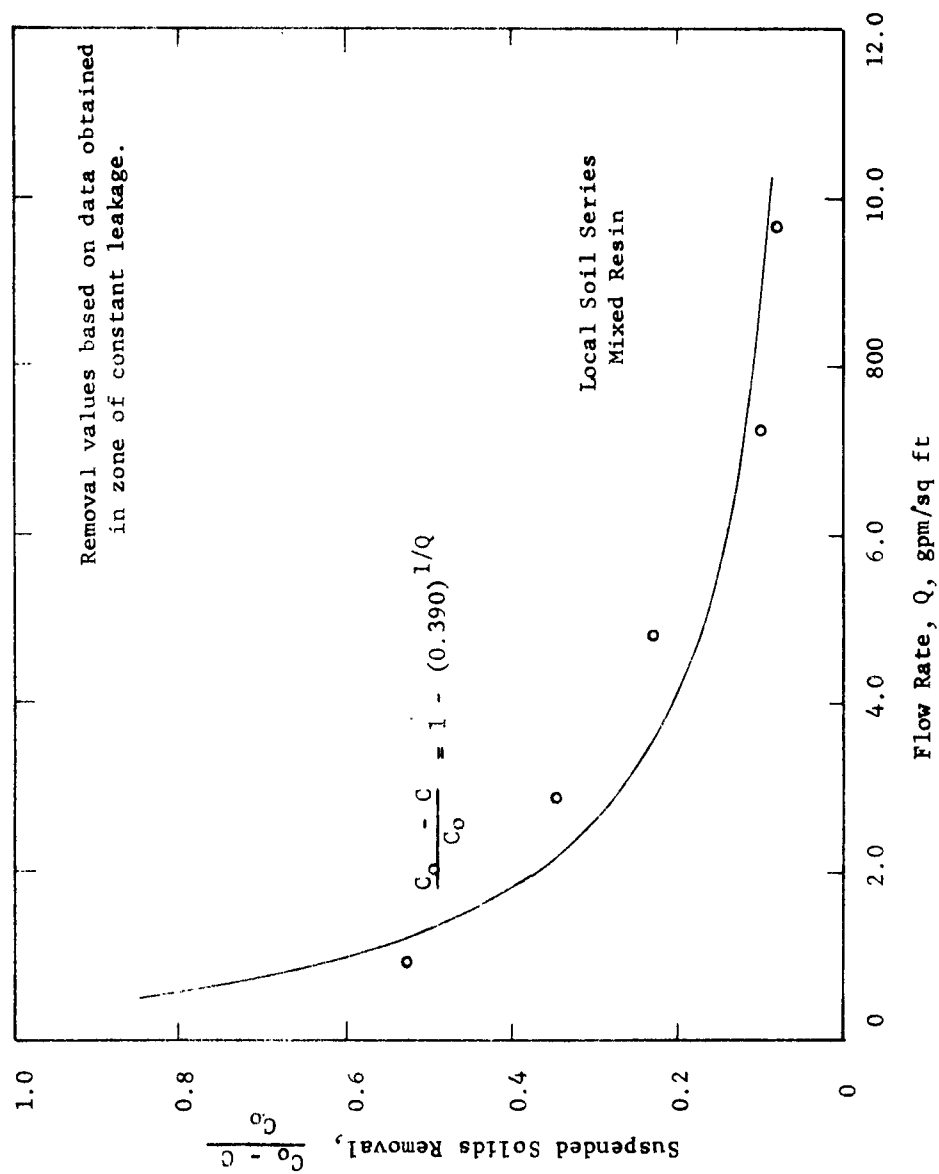
Figure 14. EFFECT OF FLOW RATE ON SUSPENDED SOLIDS REMOVAL

It may be noted that data obtained on the leakage of local soil solids through beds of mixed resin as a function of flow rate could be expressed reasonably well by equation (11). Leakage data, obtained in the zones of constant leakage, were analysed by the least squares technique in relation to the flow rates employed, and the resulting equation and curve are shown on Figure 14.

Radiostrontium Leakage in the Presence of Solids

Stock solutions of bentonite clay in distilled water were prepared and passed slowly through a bed of calcium-form cation resin in order to convert the clay to the calcium form. This procedure, previously employed (17), was reported first in this country by Lewis (54). Influent solutions were prepared by diluting the stock clay solutions with waters of the desired chemical compositions; synthetic solutions of calcium chloride were used in the present study. Radiostrontium was added to the influent solutions in the amount of about 10,000 dpm/ml, and the solutions were set aside for a period of several days to allow time for the system to approach equilibrium.

Each solution was passed through a column of sodium-form cation resin under variable flow rate conditions; the column was 1-in. diameter by 24-in. deep, and the column effluent was fractionated into 200 ml samples. Effluent was wasted at the start of each run and following each change in flow rate to minimize transient effects.

Influent and effluent samples were placed in planchets, evaporated and counted in a windowless gas flow internal proportional counter with an efficiency of about 50%.

If it is assumed that the decontamination factor, D_F, is equal to the concentration of radiostrontium in the influent divided by the effluent concentration, it may be shown (17) that the decontamination factor varies with the effective residence time, t, of the fluid within the resin bed according to

$$D_F = (D_F)_o \, e^{-kt} \tag{12}$$

where

$$t = \frac{vf}{R} \left(1 - \frac{V}{\bar{V}} \right) \tag{13}$$

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In these equations, R is the flow rate, ml/min., vf is the pore volume, ml, V is the throughput volume, ml, and $\bar{V}$ is the average breakthrough volume of radiostrontium for the given conditions. Extrapolation of experimental data for decontamination factor and effective residence time back to zero time yields the value of $(D_F)_o$; presumably this would be the decontamination factor if it were possible to achieve instantaneous and complete separation between the liquid phase and the solid phase in the influent solution.

Experimental data obtained in the present study are reported in Appendix C, and a summary of the results are presented in Table V. It may be generally conclude· that the decontamination factor for radiostrontium in the presence of suspended solid· varies directly with the ionic strength of the influent solution and inversely with the suspended solids concentration. It might be noted that previous values (17) of the dissociation rate constant k under similar conditions varied from 0.14 to 0.25 min.$^{-1}$ for radiostrontium. The difference between the two sets of data can probably be ascribed to the prolonged contact time between the radiostrontium and the suspended solids in the influent solution before passing through the resin bed in the present study, but insufficient data are available to generalize upon the variations in the values of k with specific parameters.

From the results of this study, two broad observations may be made:

    (1) Ion exchange resin beds may not be depended upon for removal of suspended solids from influent waters, although a degree of removal may be achieved in certain circumstances.

    (2) Suspended solids may carry radionuclides through a resin bed, although there is a tendency of the radionuclides to dissociate from the solids within the pores of the resin bed.

From these observations, it may be concluded that a decontamination unit for small volumes of water must incorporate filtration as well as ion exchange in order to provide a more complete degree of decontamination.

TABLE V

EXPERIMENTAL RESULTS ON RADIOSTRONTIUM LEAKAGE

	Influent Solution			Leakage Parameters	
No.	Calcium (meq/1)	Clay (mg/1)		$(D_F)_o$	k $(min.^{-1})$
1	0.2	9.5		51.	0.039
2	0.2	28.5		22.	0.037
3	0.2	95.		1.9	0.076
4	5.0	28.5		370.	0.040
5	5.0	47.5		84.	0.092
6	5.0	95.		68.	0.023

Chapter VI

SMALL SCALE DECONTAMINATION UNIT

The basis for design of a small scale decontamination unit for drinking water must be closely related to the concepts,both theoretical and practical, which have been previously discussed; in general the unit should meet the criteria of reliability, simplicity and economy in connection with the conditions which nay be reasonably anticipated following a nuclear war.

It is appropriate to review the various designs which have been proposed for small scale decontamination purposes and to discuss experimental performance results obtained with the units.

Previously Proposed Units

Lauderdale and Emmons (55) were probably the first to suggest a means for decontaminating small volumes of waters containing radioactive substances; their unit was constructed of two columns, each 3/4-in. diameter and 26-in. long, arranged in series. Water flowed upward through the first column which contained 50 g steel wool, packed to a depth of 15 inches, 37 g of calcined clay in an 8-in. layer, and 12 g of activated charcoal in a final 3-in layer. Upon leaving the first column, the water flowed downward through the second column which contained a 26-in depth of mixed synthetic organic resins. The effectiveness of the unit was investigated by experiments using reactor produced mixed fission products diluted by tap water as the influent solution, and the variables studied included flow rate and resin depth. Flow rates ranging from 0.4 to 5.0 gpm/ sq ft and resin bed depths of 13, 26, and 39 inches were employed.

It was concluded that neither removal efficiency nor total capacity of the unit was affected by the range of flow rates used, and it appeared that the total volume of water which could be treated before the effluent concentration of radioactivity exceeded a prescribed value was directly proportional to the resin

bed depth. The mixed resin bed, used alone, was not as effective as the entire unit in the removal of radioactive materials from the influent waters.

The relative amounts of radioactive material removed by the different components were reported; for example steel wool retained 85.26%, calcined clay 11.99%, activated charcoal 0.80%, cation resin 1.92% and anion resin 0.21% of the total beta emitters in the influent. It is perhaps more significant to recast these values as the percent removed from the total amount reaching each component; in this sense, the steel wool retained 85.26%, calcined clay 81.3%, activated charcoal 34.4%, and the resins removed essentially all of the remaining beta emitters. Since the unit was not loaded to saturation, it is not known whe her or not these values represent maximum amounts of uptake.

Although the steel wool had been added primarily for the removal of ruthenium, other radionuclides were removed also to some degree. The clay had been found effective in removing cesium and, to a lesser extent, strontium, but in operation it also served to filter small iron particulates originating from the steel wool. Activated charcoal was added to aid in the prevention of objectionable tastes and odors and for possible adsorption of radiocolloids. A band of rust was formed in the steel wool during the passage of water, but apparently the rust did not present operational problems.

A commercial adaptation of the Lauderdale-Emmons uint was evaluated at Oak Ridge National Laboratory in 1952 (56). This unit consisted of a homogeneous mixture of 42 g powdered iron, 36 g mixed diatomaceous earth and activated charcoal, and 44 g mixed resin packed into a single column. The resins were mixed on a weight basis which resulted in 22 g of anion resin with exchange capacity of 2.4 meq/g and 22 g of cation resin of 4.25 meq/g exchange capacity. The greater total exchange capacity of cation resin compared to that of anion resin (93.5 meq vs 52.8 meq) led to an acid effluent containing significant amounts of iron. The study concluded that the original design of Lauderdale and Emmons was superior to the commercial version. It may be noted that the observed performance of the Lauderdale-Emmons unit in this study differed

somewhat from the original study, but this was attributed to the difference between the compositions of the mixed fission products used in the two studies.

An evaluation of another commercial device for decontamination was reported by Lacy and Lindsten (57), although the original purpose of the unit was simply one of demineralization. Consisting of a 1.5-in. diameter column 6-in. long containing mixed resins. the column is fitted with a rubber adaptor for attachment to an ordinary faucet. A color indicator is incorporated with the resin, and the color changes from black to yellow upon chemical exhaustion of the resin. Mixed fission products, diluted with tap water, were applied to the column and the effluent concentration of radioactivity was measured in relation to throughput volume. Instantaneous decontamination factor values of 10^4, 10^3, and 10^2 were found at throughput volumes of 0.5, 2.6 and 10.2 liters, respectively, and the unit removed 97.1% of the influent radioactive materials after 34 liters had been treated.

The Vitro Corporation of America, under contract to the U. S. Atomic Energy Commission, developed a laboratory waste disposal unit for decontamination of liquid radioactive wastes (58). Although the unit was not specifically directed toward decontamination of emergency water supplies, results obtained during its development are of interest in the present study.

In the removal of reactor produced radionuclides from tap water, it was found that best results were obtained through the use of a mixed resin bed with the cation and anion resins in the hydrogen and hydroxyl forms, respectively. Optimum performance was observed when the two resins were present in approximately equivalent amounts, i.e., anion resin volume about twice that of the cation resin. After concluding the use of an indicating adsorbant was desirable, the investigation indicated best result were obtained with the hydroxyl form anion resin dyed pink with phenolphthalein indicator.

The unit was designed for gravity flow, and it was found that filtration prior to resin contact was desirable. A rubber sponge filter pad was selected over two forms of synthetic sponge materials on the basis of resistance to strong chemical solutions;

it appears the difference would not be significant in natural waters. During field tests of the prototype design, a problem was encountered in the tendency of the filter pad to float, but this was corrected later by supplying a restraining grid on the surface of the filter.

No benefit was derived from the substitution of activated charcoal for part of the resin bed; an additional detrimental effect was loss in the total exchange capacity of the bed. Various commercial laboratory scale demineralizers, including the one evaluated by Lacy and Lindsten (57), were tested under identical conditions. The best performance was given by the largest unit; its size was 3-in. diameter and 12-in long.

The Ionac Chemical Company of Birmingham, New Jersey, has developed a small water purifier for the purpose of removing chemical, bacteriological and radiological contaminating substances from water (59). The unit consists of a plastic treatment bag with a built-in filter, a specially formulated mixture of sorbants, and a separate supply of sterilizing tablets. Designed to operate on a batch basis, a quart of water is added to the bag containing the mixed sorbants and is drained out the bottom through the filter. Sterilizing tablets are added to the treated water, and the water is then ready for consumption. Results obtained with the treatment of one quart of water contaminated with reactor-produced radionuclides indicate decontamination factors of 145 and 335 for mixed fission products, 690 for radioiodine, 430 for radiocesium, and 1,750 for mixed fission products and radiostrontium. The unit is said to be capable of providing twenty-eight quarts of drinking water, but data on decontamination effectiveness are not available except for the first quart.

Lowe et al. (10 have shown that filtration and ion exchange may be used together as an effective means of decontamination on a small scale in tests using pulverized bomb debris. Filtration was accomplished with either a military issue hand-operated filter pad unit or a commercial silver impregnated ceramic filter candle with a hand pump. The ion exchange unit, similar to that evaluated by Lacy and Lindsten (57), was used following the filtration step.

Robeck et al. (60) suggest a unit for small scale water decontamination should contain disposable cartridges of mixed resins, the need for replacement of the cartridge may be indicated by a conductivity meter or resin color change, and a flow rate of 2 gpm/ cu ft is acceptable for operation.

It must be noted that, with the sole exception of the study reported by Lowe et al. (10), all work summarized in this section was performed with reactor produced radionuclides; extrapolation of these results to a nuclear war situation must be done cautiously because of the reported limited solubility of fallout materials.

Design Criteria for Decontamination Unit

A rational basis for design of a small-scale decontamination unit may be obtained by consideration of the nature of the problem, experimental results obtained in the laboratory and in the field, theoretical aspects, and demonstrated performance of proposed units. Specific criteria should include:

(1) Adequate provisions should be made for filtration, for ion exchange, and for sterilization of the water prior to consumption. Although these processes conceivably could be performed in a single device, it appears desirable to provide separate devices for filtration and for ion exchange

(2) The filtration unit should precede the ion exchange unit, and replaceable filter pads or elements should be incorporated in its design The filter should be capable of producing a water virtually free of turbidity, and the replacement of the filter pad or element is to be indicated by decreased flow rate through the filter.

(3) Ion exchange units will be disposable cartridges containing mixed strong acid and strong base synthetic organic exchange resins in the H-OH forms. Common commercial sizes of the resins may be used, and the total cation exchange capacity should be equivalent to the total anion exchange capacity,

i.e. the volume of anion resin should be approximately twice that of the cation resin volume before mixing.

(4) Provisions should be made for downward flow operation of the ion exchange unit; minimum size of the column of resin in 1-in. ID by 6-in. depth. The specified size is based upon hydraulic considerations, and no maximum dimensions are specified.

(5) A color indicator is to be incorporated with the resin to signify exhaustion of the resin. The column is to be constructed of rigid or semi-rigid clear plastic materials to make color observation possible.

(6) Means are to be provided for control of flow rate through the resin bed; the maximum flow rate should not exceed 10 gpm/sq ft.

(7) The final process of sterilization may be accomplished either by individual tablets or by insertion of a soluble sterilizing material at the base of the ion exchange column.

No provisions for shielding of the decontamination unit are made because of high cost in relation to need. Such ingredients as powdered metals, clay minerals and activated charcoal, although shown to be effective in decontaminating waters containing reactor-produced radionuclides, are considered to be of less total value than an equal volume of additional exchange resins.

A decontamination unit meeting the prescribed criteria would be reasonably simple in operation and would be capable of adequate decontamination performance, however it appears that the cost of the unit would be greater than desired. A detailed study of the relative costs of the decontamination unit and alternate sources of uncontaminated waters, such as emergency stores of water, is warranted under the circumstances. Preliminary estimates indicate the decontamination unit would not be more economical than stored water unless the total volume of required water exceeded two or three hundred gallons.

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APPENDIX A

EXPERIMENTAL DATA FOR LOCAL SOIL STUDIES

Run No. LS - 1

Conditions: Distilled water solutions and anion resin
Local soil concentration   500 mg/1
Flow rate   2.0 gpm/sq ft
pH   8.0

Throughput Volume (ml.)	Effluent Concentration mg/1	C/C_o	Throughput Volume (ml.)	Effluent Concentration mg/1	C/C_o
50	238	0.48	300	286	0.57
100	249	0.50	350	292	0.58
200	273	0.55	400	285	0.57

Run No. LS - 2

Conditions: Tap water solution and anion resin
Local soil concentration   500 mg/1
Flow rate   2.0 gpm/ sq ft
pH   8.0

Throughput Volume (ml.)	Effluent Concentration mg/1	C/C_o	Volume (ml.)	Effluent Concentration mg/1	C/C_o
10	2	0.00	200	35	0.07
20	3	0.01	300	31	0.06
30	78	0.16	400	31	0.06
40	96	0.19	500	49	0.10
50	79	0.16	1000	62	0.12
60	68	0.18	1500	100	0.20
70	61	0.12	2000	141	0.28
80	54	0.11	2500	210	0.42
90	51	0.10	3000	260	0.52
100	48	0.10	3500	261	0.52

APPENDIX A - continued:

Run LS-3

Conditions: Distilled water solution and mixed resin
Local soil concentration   500 mg/l
Flow rate   1.85 gpm/sq ft
pH   8.0

Throughput Volume (ml.)	Effluent Concentration mg/l	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/l	C/Co
10	0	0.00	250	425	0.85
20	0	0.00	270	440	0.88
30	3	0.01	290	439	0.88
40	20	0.04	310	440	0.88
50	31	0.06	330	450	0.90
60	41	0.08	350	446	0.89
70	78	0.16	370	444	0.89
80	133	0.27	390	450	0.90
90	184	0.39	410	446	0.89
100	249	0.50	430	454	0.91
110	278	0.56	450	471	0.94
120	322	0.64	470	460	0.92
130	353	0.71	490	454	0.91
140	385	0.77	510	456	0.91
150	406	0.81	560	454	0.91
160	408	0.82	610	454	0.91
170	419	0.84	660	462	0.92
180	425	0.85	710	468	0.94
190	420	0.84	760	456	0.91
200	438	0.88	810	458	0.92
210	420	0.84	860	463	0.93
230	420	0.84			

APPENDIX A - Continued:

Run LS-4

Conditions: Distilled water solution and cation resin
Local soil concentration   500 mg/l
Flow rate   1.75 gpm/sq ft
pH   8.0

Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co
10	1	0.00	130	491	0.98
20	0	0.00	140	495	0.99
30	77	0.15	150	493	0.99
40	26	0.05	160	491	0.98
50	131	0.26	170	493	0.99
60	239	0.48	180	495	0.99
70	360	0.72	190	491	0.98
80	420	0.84	200	493	0.99
90	475	0.95	250	475	0.95
100			300	491	0.98
110	491	0.98	350	480	0.96
120	489	0.98			

Run No. LS-5

Conditions: Distilled water solution and anion resin
Local soil concentration   500 mg/l
Flow rate   1.95 gpm/sq ft
pH   8.0

Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co
10	2	0.00	170	234	0.47
20	3	0.01	180	240	0.48
30	82	0.16	190	243	0.49
40	78	0.16	200	235	0.47
50	57	0.11	210	234	0.47
60	142	0.28	230	222	0.44
70	217	0.43	310	227	0.45
80	225	0.45	410	221	0.44
90	231	0.46	510	228	0.46
100	228	0.46	610	221	0.44
110	234	0.47	660	217	0.43
120	234	0.47	760	229	0.46
130	235	0.47	810	217	0.43
140	236	0.47	910	217	0.43
150	240	0.48	1010	211	0.42
160	235	0.47			

APPENDIX A - Continued:

Run No. LS-6

Conditions: Tap water and anion resin
Local soil concentration   500 mg/l
Flow rate   2.0 gpm/sq ft
pH   8.0

Throughput Volume (ml.)	Effluent Concentration mg/l	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/l	C/Co
0.01	2	0.00	5.0	219	0.44
0.02	3	0.01	5.5	248	0.50
0.03	78	0.16	6.0	276	0.55
0.04	96	0.19	6.5	789	1.58
0.05	79	0.16	7.0	280	0.56
0.06	68	0.14	7.5	282	0.56
0.07	61	0.12	8.0	274	0.55
0.08	54	0.11	8.5	294	0.59
0.09	51	0.10	9.0	322	0.64
0.10	48	0.10	9.5	312	0.62
0.20	35	0.07	10.0	312	0.62
0.30	31	0.06	10.5	800	1.60
0.40	31	0.06	11.0	808	1.62
0.50	48	0.10	11.5	242	0.48
1.0	62	0.12	12.0	278	0.56
1.5	100	0.20	12.5	285	0.57
2.0	141	0.28	13.0	275	0.55
2.5	210	0.42	13.5	311	0.62
3.0	260	0.52	14.0	319	0.64
3.5	261	0.52	14.5	325	0.65
4.0	953	1.91	15.0	380	0.76
4.5	301	0.60			

Run No. LS - 7

Conditions: Tap water and mixed resin
Cotton plug filter   1.88 gr., 1.37 cu. in.
Local soil concentration   250 mg./l
Flow rate   7.25 gpm/ sq ft
pH   8.0

Throughput Volume (ml.)	Effluent Concentration mg/l	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/l	C/Co
0.5	4	0.02	4.5	21	0.08
1.5	11	0.04	5.5	34	0.14
2.5	11.5	0.05	6.5	36	0.14
3.5	13	0.05	7.5	36	0.14

Appendix A - Continued:

Run No. LS-8

	Conditions:	Tap water and mixed resin	
		Local soil concentration	250 mg/1
		Flow Rate	7.25 gpm/ sq ft
		pH	8.0

Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co
0.125	88	0.35	3.250	206	0.82
0.375	143	0.57	3.750	211	0.84
0.625	163	0.65	4250	212	0.85
0.875	165	0.66	4750	212	0.85
1.250	178	0.71	5250	218	0.87
1.750	187	0.75	5750	225	0.90
2.250	198	0.79	6250	221	0.89
2.750	193	0.77	6750	225	0.90
			7250	229	0.92

APPENDIX B

EXPERIMENTAL DATA FOR STANDARD CLAY STUDIES

Run No. SC - 1

Conditions: Distilled water solution and cation resin
Kaolinite clay concentration   200 mg/l
Flow rate   1.95 gpm/sq ft
pH   6.5

Throughput Volume (ml.)	Effluent Concentration mg/l	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/l	C/Co
10	8	0.04	200	181	0.91
60	73	0.37	210	187	0.94
70	107	0.54	260	153	0.76
80	120	0.60	310	155	0.78
90	151	0.76	360	174	0.87
100	147	0.74	410	171	0.86
110	145	0.73	460	174	0.87
160	145	0.73	510	166	0.83
170	151	0.76	560	158	0.79
180	148	0.74	610	172	0.86
190	162	0.81	660	162	0.81

Run No. SC- 2

Conditions: Distilled water solution and mixed resin
Kaolinite clay concentration   200 mg/l
Flow rate   1.95 gpm/sq ft
pH   6.5

Throughput Volume (ml.)	Effluent Concentration mg/l	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/l	C/Co
50	7	0.04	360	60	0.30
60	7	0.04	380	69	0.34
70	16	0.08	390	68	0.34
80	3	0.02	400	82	0.41
90	8	0.04	410	81	0.40
100	9	0.04	420	76	0.38
150	12	0.06	430	82	0.41
250	15	0.08	480	103	0.52
300	30	0.15	530	116	0.59
310	44	0.22	540	111	0.56
320	36	0.18	590	117	0.58
330	71	0.36	640	126	0.63
340	47	0.24	690	124	0.62
350	48	0.24	700	130	0.65

APPENDIX B - Continued

Run No. SC-3

Conditions: Distilled water solution and cation resin
Illite clay concentration 250 mg/1
Flow rate 1.95 gpm/ sq ft
pH 4.5

Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co
60	89	0.36	1100	202	C.81
70	103	0.41	1200	219	0.88
80	99	0.40	1300	207	C.83
90	120	0.48	1400	211	0.84
100	122	0.49	1500	242	
260	152	0.61	1600	211	0.84
270	152	0.61	1700	219	0.88
280	141	0.56	1800	227	0.91
290	159	0.65	1900	225	0.90
300	163	0.65	2160	208	C.83
400	164	0.6	2170	208	v.83
500	177	0.71	2180	209	0.84
600	187	0.75	2190	240	
700	187	0.75	2200	216	0.86
800	207	0.83	2210	219	0.88
900	208	0.83	2220	218	0.87
1000	210	0.84	2230	233	0.93

Run No. SC-4

Conditions: 5×10^{-4} N $CaCl_2$ solution and cation resin
Illite clay concentration 250 mg/1
Flow rate 1.95 gpm/ sq ft
pH 4.5

Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co
60	170	0.68	340	230	0.92
70	189	0.75	350	242	0.97
80	225	0.90	610	250	1.00
90	192	0.77	620	250	1.00
100	205	0.82	630	242	0.97
310	235	0.94	640	250	1.00
320	237	0.95	650	250	1.00
330	144	0.98			

APPENDIX B - Continued:

Run No. SC - 5

Conditions: Distilled water solution and mixed resin
Illite clay concentration   250 mg/1
Flow rate   1.95 gpm/ sq ft
pH   4.5

Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co
60	2	0.01	900	81	0.32
70	40	0.16	1000	87	0.35
80	8	0.03	1100	108	0.43
90	22	0.09	1200	111	0.44
100	22	0.09	1300	115	0.46
310	13	0.05	1400	127	0.51
320	19	0.08	1500	135	0.54
330	22	0.09	1600	151	0.60
340	24	0.10	1660	159	0.64
350	29	0.12	1670	154	0.62
500	22	0.09	1680	155	0.62
600	52	0.21	1690	152	0.61
750	43	0.17			

Run No. SC - 6

Conditions: Distilled water and mixed resin
Bentonite clay concentration   250 mg/1
Flow rate   1.95 gpm/ sq ft
pH   7.7

Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co	Throughput Volume (ml.)	Effluent Concentration mg/1	C/Co
10	10	0.04	150	25	0.10
20	10	0.04	200	5	0.02
30	11	0.04	250	11	0.04
40	5	0.02	300	35	0.14
50	25	0.10	350	11	0.04
60	10	0.04	400	0	0.00
70	25	0.10	450	95	0.38
80	11	0.04	500	0	0.00
90	0	0.00	1000	25	0.10
100	11	0.04	1250	11	0.04

APPENDIX C

EXPERIMENTAL DATA ON RADIOSTRONTIUM LEAKAGE

Solution No. 1

Calcium   0.2 meq/1
Clay   9.5 mg/1

t (min.)	D_F
2.07	62.7
2.27	57.0
4.97	60.0
4.28	62.6
8.03	62.4
6.10	65.8
5.79	60.5
8.51	65.6
13.6	84.5
13.7	88.3
11.6	87.1
12.4	89.6
14.6	89.6
9.83	88.0
17.0	106.5
17.0	88.5
2.09	50.9

Solution No. 2

Calcium   0.2 meq/1
Clay   28.5 mg/1

t (min.)	D_F
2.46	30.2
2.49	27.4
4.47	27.1
4.49	27.1
6.06	28.6
6.10	26.2
7.65	29.7
7.60	27.9
9.27	30.5
9.27	29.1
11.70	33.3
11.73	36.3
16.06	37.1
16.06	43.7
21.63	46.6
21.55	53.0
2.61	21.5
2.63	20.9

Solution No. 3

Calcium   0.2 meq/1
Clay   95. mg/1

t (min)	D_F	(min)	D_F
13.2	5.24	2.8	2.34
13.7	5.38	2.8	2.24
13.7	5.38	2.9	2.19
13.7	5.38	2.3	2.27
13.7	5.66	5.1	2.89
5.5	2.95	6.2	2.98
6.0	2.99	5.7	2.98
5.5	3.01	4.6	3.09
5.1	3.04	8.0	3.70
4.6	2.92	7.4	3.17
2.8	2.38	8.3	3.17

Solution No. 4

Calcium   5.0 meq/1
Clay   28.5 mg/1

t (min)	D_F
2.31	1058.
4.47	1152.
5.99	346
7.40	223
9.27	319
10.6	493
12.7	727
17.5	1143
12.9	738
10.0	627
7.75	414
6.16	216
4.94	476
2.36	202
1.85	603

APPENDIX C - Continued

Solution No. 5

Calcium   5.0 meq/1
Clay   47.5 mg/1

t (min)	D_F
2.23	136.
4.19	119
5.29	121
6.52	133
8.04	144
11.3	192
12.7	282
18.6	430
12.0	374
9.61	222
7.50	182
5.97	142
4.74	124
3.85	119
2.03	97.4

Solution No. 6

Calcium   5.0 meq/1
Clay   95.0 mq/1

(min)	D_F
4.46	104.
10.1	61.8
2.63	58.7
19.1	84.1
11.2	79.0
5.88	74.7
14.1	125.
5.90	93.3
7.13	97.8
2.59	60.1
9.02	125.
1.60	53.8